Dynamics of a sliding bubble in an isothermal viscous liquid

by

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I am the author of the thesis entitled *Dynamics of a sliding bubble in an isothermal viscous liquid*

submitted for the degree of **Master of Engineering (S825)**

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Abstract

Gas-liquid flows underneath a downward facing plane play an important role in many natural and industrial processes such as spray cooling, submerged gas injection, vapour nucleation, mass transport in fluidized bed, cavitation in impeller blade and gas evolving electrodes. In particular, bubble nucleation on the electrodes in electrolysis process and its sliding characteristics are of practical interest in many industrial electrolysis processes. A bubble sliding underneath an inclined plane changes the hydrodynamics of the electrolyte by displacing the liquid, particularly in the wake region behind the bubble. In the Hall-Héroult cell, gas bubbles nucleate underneath carbon anodes under the intense magnetic field. The anodic gas bubbles have a complex effect on the electrolysis process due to the presence of several forces such as gravity, surface tension, inertia and the Lorentz force [1]. In the past decades numerous studied have been conducted on the bubble-induced flow in the aluminium reduction cell with bench-scale physical analogue models [2-5], as experiments on an actual operating cell are impractical. In most of these laboratory-based models, the bubbles are generated by injected air through a submerged orifice or forcing air through a porous plate, which differs considerably to the Hall-Héroult process, where the gas bubbles are generated by electrolysis under a strong external magnetic field. Despite the efforts made towards the development of lab-based physical model [6], there still remains a number of concerns about the influence of a magnetic field on gas bubbles. These issues provide the motivation to study the effect of an external magnetic field on the gas-liquid interaction in the electrolysis process.

The primary objective of this research is to study the influence of a static external magnetic field on electrolytically generated bubbles in a lab-scaled aqueous electrolytic cell. The research work is divided into two parts, reflecting the chronology of the experimental work performed. The first part presents a study of the most important hydrodynamic characteristics of bubble induced flow in the aqueous electrolytic media. Bubbles were generated using aqueous copper sulphate (CuSO₄) solution of 0.5 molar concentration by maintaining 3 amperes (800 A/m²) of total current from a DC power source. Photographic techniques were extensively used to quantify the bubble sliding characteristics under the influence of an external magnetic field. Experiments were carried out at different inclination angles of the anode (θ = 0°, 1° and 2°) with and without the magnetic field. ImageJ was used to measure the bubbles sliding velocities, and their sliding-paths were plotted by manual tracking, based on the maximum pixel intensity. Most of the bubbles were found to move in a cluster (or in a bubble swarm), exhibiting a sluggish flow at some parts of anode surface. A significant finding was the enhancement of the bubble velocity due to the coalescence process in the presence of
an external magnetic field. However, it was rare (difficult) to see any single bubble sliding across the anode due to the smaller size of the nucleated bubbles, the bubble swarm and the bubble-bubble interaction.

In the second part, the physical model was modified with an air-injection system to create a single bubble in electrolytic solution to study the influence of magnetic field on a single/isolated bubble. As the velocity of the single bubble deviates significantly from the linear trajectory in presence of a magnetic field, simultaneous velocity measurements are required to estimate the relative velocities between them. A particle image velocimetry (PIV) study was conducted to measure the velocity of the surrounding fluid, with and without the magnetic field. The bubble tends to form a rotational path due to the combined effect of buoyancy and MHD fields. The instantaneous bath velocity vector maps were correlated with the dimensionless numbers. The results showed more realistic hydrodynamic characteristics due to the presence of a static external magnetic field.
Nomenclature

\[ \eta_{\text{act}} \quad \text{Activation Polarization} \]
\[ R_T \quad \text{Total Resistance} \]
\[ R_{\text{BFZ}} \quad \text{Resistance in the bubble-free zone} \]
\[ R_{\text{BLZ}} \quad \text{Resistance in the bubble layer zone} \]
\[ L_{\text{ACD}} \quad \text{Anode-to-cathode distance} \]
\[ h \quad \text{Bubble layer thickness} \]
\[ A_{\text{a}} \quad \text{Conductive surface area of the anode} \]
\[ \kappa_0 \quad \text{Electrical conductivity of the bubble-free zone} \]
\[ \kappa_B \quad \text{Electrical conductivity of the bubble layer conductivity} \]
\[ \varepsilon \quad \text{Volume fraction of the gas phase} \]
\[ \Theta \quad \text{Coverage factor} \]
\[ A_t \quad \text{Total projected bubble area} \]
\[ d_B \quad \text{Diameter of large bubbles} \]
\[ q \quad \text{Gas generation rate per unit surface area} \]
\[ g \quad \text{Acceleration due to gravity} \]
\[ \rho_1 \quad \text{Liquid density} \]
\[ \rho_g \text{ and } \rho_b \quad \text{Gas density} \]
\[ \sigma \quad \text{Surface tension} \]
\[ L \quad \text{Characteristic dimension} \]
\[ \mu_1 \quad \text{Viscosity of surrounding liquid} \]
\[ F_b \quad \text{Buoyancy force} \]
\[ F_d \quad \text{Drag force} \]
\[ J \quad \text{Current density vector} \]
\[ B \quad \text{Magnetic field vector} \]
\[ \dot{F} \quad \text{Volumetric source term} \]
\[ \rho_p \quad \text{Tracer particle density} \]
\[ U_g \quad \text{Gravitational induced velocity} \]
\[ d_p \quad \text{Particle density} \]
\[ U_p \quad \text{Velocity of the particle} \]
\[ K_p \quad \text{Distance between the adjacent pixel} \]
\[ V_b \quad \text{Velocity of the bubble} \]
\[ \varphi \quad \text{Bubble coverage} \]
\[ \theta \quad \text{Anode inclination} \]
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Introduction

Ever since the invention of the Hall-Héroult process in 1886, it has remained as the main process of producing aluminium by alumina even with the significant advances in technology during the past century. One of the main concerns of this process has been the high energy requirement, approximately 13 kWh for each 1kg of aluminium produced, and of that only half of the energy is actually utilized to decompose alumina into aluminium. Meanwhile, the rest is lost as waste heat. The financial cost and environmental consequences have lead the aluminium industry into reducing its energy consumption [7, 8].

Over the last few decades, many refinements and considerable developments has been made to the Hall-Héroult cell. Even with these advancements, the aluminium industry is constantly pursuing further gains in productivity and efficiency, whilst lowering the consumption of energy [9]. Recent studies into the effect of MHD induced flow have shown the significance of this source of bath momentum in the cell process with positives and negatives. Despite the recent interest in the MHD induced flow, its effects on the bubbles that are formed underneath the anode are not well understood. A major reason behind this is due to the fact that acquiring data from an industrial (operating) cell is difficult. This also limits the data for the validation of Computational fluid dynamics (CFD) models. Thus, many have utilized physical models, however, no one has yet been able to design a model that incorporates bubble formation with MHD forces.

Previous studies that reported on the characteristics of bubbles sliding under walls in the Hall-Héroult cells have mainly used pressurized air to generate air bubbles under surfaces with no magnetic field. They either assumed the effects of MHD forces as negligible or ignore MHD as a source of momentum in the bath. More recent studies have showed that MHD forces contribute to the bath velocity, hence, this may affect the bubble removal process [10-12]. The aim of this project is to identify and contrast the effects of magnetic field on the behaviour of bubbles under an inclined surface. In Chapter 2, the bubbles are generated by an electrolytic process similar to that of the Hall-Héroult process. This chapter will focus on the effect of MHD forces on the bubble motion in isothermal conditions.

As it is nearly impossible to simulate a Hall-Héroult process in laboratory conditions, certain assumptions have been made. Firstly, the chosen electrolyte is an aqueous copper (II) sulphate solution instead of the traditional complex bath composition of alumina, cryolite, etc. This simplification is necessary to run the electrolysis process in laboratory conditions. Secondly, the amount of electricity used is only up to 3A (800 A/m²), in contrast to 180-500kA of an
industrial cell. This is an important change due to the electrical hazard and in relativity to the scale of the model. Thirdly, the experiment is kept isothermal and operated under room temperature and pressure. An operating industrial cell is maintained at 950 °C with a corrosive molten bath [13]. Finally, the Lorentz force will affect the electrolyte itself, in contrast, to the aluminium metal deposit and cryolite, in the actual aluminium reduction cell.

The air-injection modelling method that has been widely used requires attentive validation with experimental data to approach the larger challenge [13]. Nevertheless, the air-injection model can avoid the swarm of bubbles and focus on the fundamental effects of bubble behaviour. Therefore, in Chapter 3 an air-injection physical model with MHD forces is studied since it is helpful in understanding the fundamental effects. However, the single bubble model does not represent the realistic generation and type of bubbles generated in an industrial electrolysis process. Therefore, the results of physical models may not be able to be directly applied to an industrial cell [6, 8]. In Chapter 4, a particle image velocimetry (PIV) study will provide quantitative insight into the flow behaviour induced by the sliding bubble and the flow induced by magnetic field. The PIV measurements will also provide necessary validation of the dominance of MHD flow field and if these two momentum sources are complementary in the resulting flow field.
Chapter 1

Literature Review

1.1 Introduction

Bauxite is the most abundant mineral in the earth’s crust. It consists of about 50 per cent alumina (Al₂O₃) and is generated by weathering of sedimentary rocks with a high proportion of aluminium containing minerals [14]. Specifically, the primary minerals that make up bauxite are gibbsite (Al₂O₃·3H₂O), boehmite (Al₂O₃·H₂O) and diaspora (similar composition to boehmite, but with a higher density and hardness) [15]. Bauxite is refined by the Bayer process to extract the alumina. The aluminium and oxygen in alumina is separated by the smelting process to generate aluminium. Typically, one tonne of aluminium requires two tonnes of alumina to be processed [14].

In 1825, Hans Christian Oersted produced a small quantity of aluminium with heated potassium amalgam with aluminium chloride. Twenty years prior to the experimentation by Hans Christian Oersted and Sir Humphrey Davy named the produced metal as “aluminum” [16].

In the 1880’s, Charles Martin Hall developed a commercial extraction process for aluminium with electricity. Also, within few weeks Paul Héroult from France independently created a similar process. Over the years, many energy improvement have been made to the Hall-Héroult process, such as, computerization of smelters, improvements in batch chemistry, insulation, baking technology for carbon anodes and reduction in anode consumption [16].

Aluminium is widely used in the transport, construction, packaging and electrical industries. In the transport sector, aluminium is present in numerous components of cars (engine blocks, cylinder heads, transmission housings and body panels), trucks, buses, railway stock and aircraft. In construction, sheet products (roofing and wall cladding), extrusions for windows and doors, casting on hardware used by builders are made using aluminium. In packaging, aluminium alloy sheets are used for beverage can components, foils (household and commercial) and in cartons for fruit juice and pharmaceutical packaging. Also, in the electrical sector, aluminium is present in wires, generally with steel to form cables [17].
1.2 **Hall-Héroult Process**

The Hall-Héroult cell or aluminium reduction cell (shown in Figure 1) is the primary method of producing aluminium by alumina that involves electrochemical reactions, hydrodynamics due to formed gases, MHD forces, and complex heat transfer [18]. The bath consists of alumina (Al₂O₃) that is dissolved in molten cryolite (Na₃AlF₆) at a temperature of approximately 950°C (1223K). The solution is reduced to liquid aluminium that deposits at the bottom of the cell due to its marginally higher density than that of the bath [6]. The aluminium that is produced is siphoned out of the cell at certain intervals of operation [19]. The oxygen ions that are generated at the anode react with the carbon of the anode to form carbon dioxide. The following balanced equation is the overall simplified reaction [6]:

\[ 2\text{Al}_2\text{O}_3 + 3\text{C} = 4\text{Al} + 3\text{CO}_2 \]  

**Figure 1.** Cross sectional view of an aluminium reduction cell

Cryolite is a poor conductor of electricity and a large proportion of the supplied energy is released in the bath as joule heating (resistive heating). Therefore, to reduce this form of energy loss, the bath is kept at the minimum level [19, 20]. This distance is known as the anode-metal distance. The aluminium deposit below the bath is a highly conducting fluid that directly connects to the cathode at the bottom of the cell. Generally, the anode-metal distance is in the order of few centimetres vertically, in comparison to the horizontal displacement that is in the order of several meters. The primary factor that causes difficulty in reducing the anode-metal distance is the instability of the conducting fluid caused by the Lorentz force. The magnetic field present is in the order of 0.01 Telsa and the flow velocity can be typically at 0.2m/s. This velocity causes movement of the interface between the two fluids which leads to instabilities in the process. The anode-metal distance therefore must by small to reduce the energy cost and
large enough to avoid short-circuits. In a case where the interface touches the anode, the electrical hazard is substantial, especially with the large amount of power involved. As each cell of a potline is connected to each other, there is a possibility of destroying other connected cells by a short circuit from single cell [20, 21]. Furthermore, both vertical motion in interface waves and turbulence can result in the reoxidation of aluminium deposit [22].

1.3 Electrolysis Reaction

The redox reaction occurs at the bath-metal interface [21].

\[ \text{Al}_2\text{O}_3 + 6e^- \rightarrow 2\text{Al}^{3+} + 3\text{O}^{2-} + 6e^- \rightarrow 2\text{Al} + 3\text{O}^{2-} \]  

eq. 2

At the anode the following reaction takes place, where the oxygen ions react with the carbon on the anode to form carbon dioxide gas at the anode [21]:

\[ 3\text{O}^{2-} + \frac{3}{2}C \rightarrow \frac{3}{2}\text{CO}_2 + 6e^- \]  

eq. 3

Other than the ideal reactions that are expected, some undesirable reaction also occurs in the cell, which result in loss of current efficiency. The formed aluminium metal may react with some components of the bath, the following are the main reactions that dissolve the metal [23].

\[ 3\text{NaF} + \text{Al} \leftrightarrow 3\text{Na} + \text{AlF}_3 \]  

eq. 4

\[ \text{AlF}_3 + 2\text{Al} \leftrightarrow 3\text{AlF} \]  

eq. 5

The sodium that is produced in the equilibrium in eq. 4 dissolves in the aluminium metal and the bath. The sodium concentration rises with the increase of NaF:AlF\textsubscript{3} ratio in the aluminium-bath interface. The equilibrium on eq. 5 generates an interfacial concentration of monovalent aluminium species. The concentration of these species increases when the NaF:AlF\textsubscript{3} ratio decreases. The reactions on eq. 4 and eq. 5 move to the right with rising temperature generating higher interfacial concentrations that lead to a lower current efficiency. As sodium is the dominant dissolved metal, a low NaF:AlF\textsubscript{3} ratio encourages higher current efficiencies. The following reactions oxidize the sodium and aluminium monofluoride [23]:

\[ 6\text{Na} + 3\text{CO}_2(g) + 2\text{AlF}_3 \rightarrow \text{Al}_2\text{O}_3 + 6\text{NaF} + 3\text{CO}(g) \]  

eq. 6

\[ 3\text{AlF} + 3\text{CO}_2(g) \rightarrow \text{AlF}_3 + \text{Al}_2\text{O}_3 + 3\text{CO}(g) \]  

eq. 7
In order for the reactions from eq. 4 to eq. 7 to take place, the aluminium fluoride must diffuse through the cathode boundary layer with the aluminium monofluoride and dissolved aluminium must diffuse out. The Na\(^+\) traverse due to current but instead of Na\(^+\) discharging the AlF\(_3\) discharges at lower potential. This results in a higher NaF:AlF\(_3\) ratio at the bath-metal interface than the bulk and a thicker boundary layer reduces the diffusion of metal out but it reduces the mass transport that increases the NaF and Na concentrations at the bath-metal interface. On a given boundary layer thickness, the area of the cathode is proportional to the completed amount of dissolved metal diffusing out. The current efficiency can be improved by a high current density at the cathode and more aluminium is produced in comparison to the amount of dissolved metal that enters the bath for reoxidation [23].

1.4 Electrolyte Composition

The electrolyte has four main functions in the Hall-Héroult process [24]:

- Act as the solvent for the alumina in the electrolytic decomposition that results in molten aluminium and carbon dioxide.
- Medium for the electricity to pass from the anode to the cathode.
- Physically separate the aluminium metal formed at the cathode from the carbon dioxide gas at the anode.
- Acts as a heat-generating resistor that makes the cell self-heating.

The cryolite accounts for 75 to 80 wt% of the electrolyte and generally is comprised of aluminium fluoride, calcium fluoride, and alumina. These components of the electrolyte lower the melting point, the cell operating temperature and raise the efficiency of the electrolysis process. The melting point of alumina is over 2050°C and it is a chemically stable compound. The alumina that is used in aluminium reduction cells is not 100% pure and contains small amounts of oxides, such as, Na\(_2\)O, CaO, Fe\(_2\)O\(_3\), and SiO\(_2\) [24].

The composition of alumina plays a crucial role in the efficiency of the aluminium reduction process. However, a high composition of alumina in the bath due to overfeeding can lead to formation of “sludge” or “muck” under the aluminium pad. The upper limit for the alumina concentration in the bath is around 4% and over this limit the alumina halts dissolving at an adequate pace which leads to undissolved alumina settling on top of the cathode. This formation decreases the electrical conductivity of the cell and results in non-uniform current distribution, resulting in a condition called “sick pot” or “sick cell”. Moreover, the undissolved
alumina may support erosion effects because alumina is extremely abrasive and scours when coupled with the magnetohydrodynamics forces of the aluminium deposit. Thus, the cathode life is considerably reduced [25]. The decrease in alumina concentration in the bath can lead to a phenomenon called the “anode effect”, which usually occurs at a range of 1.5% to 2% alumina concentration. A voltage drop of 4 to 4.5 volts in a cell can reach up to 15-30 volts due to the anode effect. The manual termination of the anode effect takes around 5 to 10 minutes, however, to bring a cell back to normal operating conditions takes a significant amount of time. Hence, the general industrial guideline is to maintain alumina concentration at the lower part of the concentration range to avoid overfeeding [26].

The exact concentration of alumina at the point of anode effect depends also on the critical current density at the anode, temperature and composition of electrolyte. However, the anode effect arises at an alumina concentration below 2% by electrolyte weight. The anode effect diminishes the productivity of the cell and requires more power for the operation of the cell. Furthermore, this leads to the production of perfluorocarbons (PFC’s), hydrogen fluoride (HF) and other gaseous fluorinated products. Emission of PFCs and HF gases has serious consequence on the environment. The ideal concentration of alumina that is maintained in industry is within the narrow window of 2-4% alumina concentration [25].

Aluminium fluoride (AlF₃) can account for 9 to 12 wt% of the electrolyte. Three mechanisms in the cell consumes the aluminium fluoride. Firstly, it reacts with sodium oxide that is present in the impurities in the alumina. The reacted aluminium fluoride must be replaced to maintain its concentration in the electrolyte. Generally, there is about 20kg of AlF₃ per metric ton of Al produced. Secondly, AlF₃ can be depleted by hydrolysis due to the moisture (eq. 8). The hydrogen fluoride that is formed is extremely hazardous, however, the improved efficiencies of gas scrubbing and fume capture in aluminium smelters have been able to reduce the emission of hydrogen fluoride to the potroom [24].

\[
2\text{AlF}_3 + 3\text{H}_2\text{O}(g) = \text{Al}_2\text{O}_3 + 6\text{HF}(g)
\]  

eq. 8

Lastly, AlF₃ is lost in the form of vapour off the electrolyte, for example, sodium tetrafluoroaluminate vapour (NaAlF₄). Over 98% of fluorides generated are contained by the cleaning process at the fume-treatment plant and are returned to the cell with secondary alumina [24]. Calcium fluoride is added to the electrolyte due to the minor impurity of calcium oxide, at around 0.035 wt%. This results in a steady-state concentration of 4-7 wt% of calcium fluoride in the melt. From this a minor quantity of calcium is codeposited into the aluminium with some emitted as a calcium compound [24].
The complicated chemical composition and their effects on the cell process cannot be adequately modelled in a simplified and scaled down physical cell where it is necessary to maintain simplicity and ability to be safely operated in a laboratory. The electrolyte composition is an important component for both the industrial cell and the physical model and their differences and similarities have to be thoroughly understood and taken into account.

1.5 Electrical Energy Requirements

The electrical current that is required by the Hall–Héroult process is in the order of $10^5$ A [21]. Globally, the DC current for a large commercial cells is in the range of 350 – 500kA [9, 19]. The most recent aluminium smelters require around 13kWh to generate 1 kg of aluminium. On average, the DC current consumption is almost 14kWh/kg. Generally, the cost of energy accounts for 30% of the total aluminium production cost. The energy requirement for aluminium production has reduced over the years due to advances in technological improvements of the production process. At the same time the global demand for electrical energy has been steadily increasing [24].

According to World Aluminium and International Aluminium Institute [27] up to 60% of the energy consumed in the Hall-Héroult process globally is generated by fossil fuel. The environmental impact through global warming, consumption of non-renewable and renewable energy sources and depletion potential of abiotic resources is a serious concern for the aluminium production industry. Therefore, the energy consumption has to further reduce to avoid these environmental risks and to reduce the financial costs.

1.6 Busbar

A smelter has many cells that are connected transversely in rows to form a potline, and on an aluminium smelter there can be more than one potline. This connection ensures that the DC current from one cell is transferred to the other cell and so forth along the potline. The current is passed from an upstream (US) cell cathode bus to the endriser and then to the anode-bus. The horizontal busbars carry out the current from the cathode outwards to the next downstream (DS) cell [12, 28, 29]. Figure 2(a) identifies the bus system of a convectional cell and Figure 2(b) shows the current traversing in this system. The high current in the cathode busbars generates strong magnetic fields [9].

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A schematic of a 140kA potline is shown in Figure 3. The two sides of the cell are defined as tap-end (TE) and duct-end (ED). In terms of the magnetic fields expected, AB and EF line are expected to have a no symmetrical magnetic field due to the one adjacent potline. The CD line, however, has two potlines on the either side of the TE and ED, hence, a symmetric magnetic field is present in the CD line. Furthermore, the magnetic field from the adjacent line can be neglected since it is inversely proportional to the distance between the potlines (about 32m). The high intensity magnetic fields that the cathode busbar in this complex system influences positive and negative effects in the cell in terms of the MHD forces. In practice, the configuration and retrofits of an aluminium reduction plant, such as extra side, risers can change the magnetic field that is present in the cell [12].

Figure 2. One half of a convectional side-to-side endriser cell (a) mesh model (b) CAD model with current direction [12]
1.7 Anodic Gas

The formation of gas bubbles at the anode is a by-product of the Hall-Héroult process. The oxygen ion produced in the electrolysis of alumina forms carbon dioxide (eq. 3) with the carbon of the anode. The bubbles are generated at preferential sites called “nucleation sites” and once they grow to a critical volume the bubbles start to move towards the edges and escape from the anode along the vertical sidewall until they escape the bath through the air-bath interface. The presence of gas bubbles has both positive and negative effects on the Hall-Héroult process [30].

On one hand, the produced anodic gas bubbles are a significant source of energy consumption in the Hall-Héroult process. The gas bubbles produced at the underside of the anode act as an insulator that separates the anode surface from the bath liquid [6, 7]. The bubbles have a

Figure 3. Schematic of a 140kA potline [12]
complex effect on the overall electrical resistance of the cell [8]. According to Haupin [31], the bubbles generate a voltage drop in the order of 0.25 V out of total cell voltage of 4 - 4.6 V. The increase of the ohmic overvoltage of the cell can be a factor in destabilizing the operation of the cell by the unsettling of bath-metal interface and even engendering the anode effect [22]. The anode effect occurring in cells causes the generation of potential greenhouse gases and consumes higher electrical energy. The released gases can vary from the CO$_2$(g) and CO(g) to certain perfluorocarbon (PFC) compounds, such as, CF$_4$(g) and C$_2$F$_6$. These gases are classified as greenhouse gases (GHG) with high global-warming potential and atmospheric life spans in the order of 10,000 years. Carbon monoxide accounts for 70-80% of the evolved anode gas [24]. These emissions can be lowered by lowering the occurrences of anode effects per cell per day (also known as, anode effect frequency) and the anode effect duration (in minutes). The modern prebaked cells can operate for long periods of time without the occurrence of an anode effect. In some circumstances the termination of an anode effect requires manual interaction by an operator. The released CO(g) in the atmosphere in the potroom may be harmful to an operator in such circumstances [24, 32]. In 2011, the GHG emissions for a ton of aluminium produced was 1.6 tonnes of CO$_2$ and 0.6 tonnes of CO$_2$ equivalent of PFC’s [27].

On the other hand, the generated bubbles homogenise the alumina distribution and the temperature field [6, 22]. The horizontal transport of mass and energy is vital for an optimized operation. This convection is generated by the bubbles and MHD forces. The bubbles transforms the gravitation energy into the horizontal kinetic energy [22]. The bubbles add momentum to the bath via drag forces [4]. The motion of bubbles not only improves the mixing of the bath due to the increase in turbulence intensity but also the dissolution rate of alumina in the bath. The quasi-horizontal flow below the anode that is induced due to bubbles encourages the direction of heat flux towards the side wall, homogenising the temperature field [30]. The movement of bubbles improves the alumina dispersion in local areas. In addition, the vertical dispersion rate is improved in the initial stage of mixing [10].

1.7.1 Classical Bubble Resistance

The classical bubble resistance model is defined by two separate resistances. Firstly, the bubble layer zone (BLZ) with a thickness of $h$. Secondly, the bubble-free zone (BFZ) with a thickness of $L_{ACD} - h$ (Figure 4). Therefore, the total resistance ($R_T$) is calculated as shown below [30]:
Figure 4. Inter-electrode zone with the parameters used to calculate the classical bubble resistance

Where $L_{ACD}$ is the anode-to-cathode distance (ACD), $h$ is the bubble layer height, $A_a$ is the conductive surface area of the anode, electrical conductivity in the bubble-free zone is $\kappa_0$, and bubble layer zone conductivity is $\kappa_B$. The relative resistance of eq. 9 is given as shown in eq. 10 [30].

$$\frac{R_T}{R_0} = \frac{h}{L_{ACD}} \left( \frac{\kappa_0}{\kappa_B} - 1 \right) + 1$$  \hspace{1cm} \text{eq. 10}$$

The following equation by Bruggeman [33] relates the two conductivities $\kappa_0$ and $\kappa_B$. Where the volume fraction of the gas phase is given as $\varepsilon$.

$$\frac{\kappa_0}{\kappa_B} = (1 - \varepsilon)^{-3/2}$$  \hspace{1cm} \text{eq. 11}$$

The relation in eq. 11 is only valid in cases with small void fraction and objects that are uniformly distributed. The general classical model is obtained by substituting the eq. 10 into eq. 11 as given below [30]:

$$\frac{R_T}{R_0} = \frac{h}{L_{ACD}} \left( (1 - \varepsilon)^{-3/2} - 1 \right) + 1$$  \hspace{1cm} \text{eq. 12}$$

Hyde and Welch [34] used ceramic objects of known volume and shape to simulate bubbles in an electrolytic cell that produces lead. The results showed that the bubble resistance was
mainly dependent on its volume with small effect due to the shape of the bubble. Therefore, they proposed the relation given below.

\[
\frac{R_T}{R_0} = \frac{1}{L_{ACD}} \left[ (L_{ACD} - h) + \frac{h}{1 - \Theta} \right]
\]

eq. 13

The Coverage factor (\(\Theta\)) is the total projected bubble area on anode (\(A_t\)) divided by the anode area (\(A_a\)) [30].

\[
\Theta = \frac{A_t}{A_a}
\]

eq. 14

Thonstad, et al. [35] developed an expression (eq. 15) that contains the perturbations caused by the existence of bubbles, for instance, the screening effect and the electrical field deformation in the BFZ. The diameter of large bubbles is given as \(d_B\). An incremental voltage drop due the bubbles can be up to 30\% higher than the voltage drop calculated by the classical models [30].

\[
\frac{R_T}{R_0} = 1 + \left( \frac{1}{1 - \Theta} - 1 \right) \left( \frac{h}{L_{ACD}} + \frac{1 - \frac{h}{L_{ACD}}}{1 + 5\sqrt{\Theta} \frac{2(L_{ACD} - h)}{d_B}} \right)
\]

eq. 15

1.7.2 Complex bubble-induced resistance

The amount of gas holdup under the anode is time dependent and this phenomenon is termed in chemical engineering as the “gas-holdup rate”. The coverage factor can be used as an indication of the rise of resistance due to the existence of bubbles under the anode. The screening effect is one of the mechanisms that bubbles contribute to electrical resistance in the cell. This is caused by the reduction of electrical conductive area of the anode by presence of bubbles. Also, bubbles can deform the homogeneous current density field and alternate the balance of current output from the bottom and the sidewall of an anode [22, 30]. Zoric and Solheim [36] were the first to report on the deformation of the electric current field in the anode-cathode region due to bubbles under the anode. The main component of electrical resistance by large gas bubbles is due to the distortion of the current path while the partial restriction of the anode area is less significant [36]. A proportion of the overvoltage accounts for the activation polarization. It is necessary to overcome the activation energy of the reaction.
The activation polarization can be express as eq. 16, with ‘a’ and ‘b’ being Tafel constants [8].

\[ \eta_{act} = a + b \log \left( \frac{1}{A} \right) \]  

Eq. 16

The current density plays an important role on the behaviour of anodic gas bubbles, where the size of bubbles, thickness of bubble front, average fraction of the anode covered by bubbles and the bubble velocity increases with increased current density [37]. Recently, Zhang, et al. [7] assessed the bubble-induced electrical resistance in the aluminium reduction cell with a physical air-water model and a 2D numerical model. The numerical model predicted the current flow and voltage drop across the electrolytic cell with and without bubbles. This study investigated the effect of bubble coverage, effect of bubble size, effect of a large bubble and the effect of bubble thickness (Table I).

**Table I. Bubble simulation case information [7]**

<table>
<thead>
<tr>
<th>Simulation cases</th>
<th>Gas flow rate (L min(^{-1}))</th>
<th>Anode bubble coverage (%)</th>
<th>Number of bubbles</th>
<th>Bubble size (mm)</th>
<th>Bubble thickness (mm)</th>
<th>Purpose</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>4</td>
<td>37</td>
<td>20</td>
<td>Varied</td>
<td>5</td>
<td>Effect of bubble coverage</td>
</tr>
<tr>
<td>B</td>
<td>4</td>
<td>40</td>
<td>22</td>
<td>Varied</td>
<td>5</td>
<td>Effect of bubble coverage</td>
</tr>
<tr>
<td>C</td>
<td>4</td>
<td>36</td>
<td>18</td>
<td>Varied</td>
<td>5</td>
<td>Effect of bubble coverage</td>
</tr>
<tr>
<td>D</td>
<td>8</td>
<td>50</td>
<td>19</td>
<td>Varied</td>
<td>5</td>
<td>Effect of bubble coverage</td>
</tr>
<tr>
<td>E</td>
<td>8</td>
<td>47</td>
<td>13</td>
<td>Varied</td>
<td>5</td>
<td>Effect of bubble coverage</td>
</tr>
<tr>
<td>F</td>
<td>8</td>
<td>46</td>
<td>17</td>
<td>Varied</td>
<td>5</td>
<td>Effect of bubble coverage</td>
</tr>
<tr>
<td>G</td>
<td>–</td>
<td>0</td>
<td>0</td>
<td>–</td>
<td>–</td>
<td>Base case</td>
</tr>
<tr>
<td>H</td>
<td>–</td>
<td>50</td>
<td>5</td>
<td>65</td>
<td>5</td>
<td>Effect of bubble size</td>
</tr>
<tr>
<td>I</td>
<td>–</td>
<td>50</td>
<td>10</td>
<td>32.5</td>
<td>5</td>
<td>Effect of bubble size</td>
</tr>
<tr>
<td>J</td>
<td>–</td>
<td>50</td>
<td>20</td>
<td>16.25</td>
<td>5</td>
<td>Effect of bubble size</td>
</tr>
<tr>
<td>K</td>
<td>–</td>
<td>100</td>
<td>1</td>
<td>650</td>
<td>5</td>
<td>Effect of very large bubbles</td>
</tr>
<tr>
<td>L</td>
<td>–</td>
<td>50</td>
<td>20</td>
<td>16.25</td>
<td>3</td>
<td>Effect of bubble thickness</td>
</tr>
<tr>
<td>M</td>
<td>–</td>
<td>50</td>
<td>20</td>
<td>16.25</td>
<td>4</td>
<td>Effect of bubble thickness</td>
</tr>
<tr>
<td>N</td>
<td>–</td>
<td>50</td>
<td>20</td>
<td>16.25</td>
<td>6</td>
<td>Effect of bubble thickness</td>
</tr>
<tr>
<td>O</td>
<td>–</td>
<td>50</td>
<td>20</td>
<td>16.25</td>
<td>7</td>
<td>Effect of bubble thickness</td>
</tr>
</tbody>
</table>

They predicted a uniform current flow (shown in Figure 5) in the anode, ACD regions and the side channel with no bubbles. The predicted current flow with the 37% bubble coverage is shown in Figure 6. The overall current flow is similar in both cases, hence, the presence of bubbles does not impact the overall current flow in the cell. The close-up view of the no bubble simulation shows a uniform distribution of current in the anode-bath interface (Figure 5(b)). However, due to the high electrical resistance of the gas bubble the current does not pass through the bubble, instead, the current flows through the gaps between the bubbles (Figure 6(b)). As a result, at the intersecting point of bubble, anode and the interface, a point of high local current density concentration can be seen. This results in an additional voltage drop which is considered as the bubble-induced voltage drop. The predicted voltage drop and
bubble-induced resistance increased with increase in bubble coverage (Table II). An exception in this trend is case D which had a higher bubble coverage than case E, but the predicted voltage drop was lower. However, the number of bubbles is higher in case D, which results in smaller bubbles [7].

Table II. Bubble-induced resistance and voltage drop for each case [7]

<table>
<thead>
<tr>
<th>Simulation cases</th>
<th>Bubble-induced resistance [ohm]</th>
<th>Bubble-induced voltage drop [v]</th>
<th>Total voltage drop [v]</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.000248</td>
<td>0.143</td>
<td>1.532</td>
</tr>
<tr>
<td>B</td>
<td>0.000286</td>
<td>0.160</td>
<td>1.549</td>
</tr>
<tr>
<td>C</td>
<td>0.000233</td>
<td>0.137</td>
<td>1.526</td>
</tr>
<tr>
<td>D</td>
<td>0.000590</td>
<td>0.313</td>
<td>1.702</td>
</tr>
<tr>
<td>E</td>
<td>0.000640</td>
<td>0.323</td>
<td>1.712</td>
</tr>
<tr>
<td>F</td>
<td>0.000578</td>
<td>0.298</td>
<td>1.687</td>
</tr>
<tr>
<td>G</td>
<td>0</td>
<td>0</td>
<td>1.393</td>
</tr>
<tr>
<td>H</td>
<td>0.001123</td>
<td>0.511</td>
<td>1.9</td>
</tr>
<tr>
<td>I</td>
<td>0.000648</td>
<td>0.311</td>
<td>1.7</td>
</tr>
<tr>
<td>J</td>
<td>0.000426</td>
<td>0.178</td>
<td>1.567</td>
</tr>
<tr>
<td>K</td>
<td>0.034719</td>
<td>11.247</td>
<td>12.636</td>
</tr>
<tr>
<td>L</td>
<td>0.000363</td>
<td>0.157</td>
<td>1.546</td>
</tr>
<tr>
<td>M</td>
<td>0.000911</td>
<td>0.170</td>
<td>1.559</td>
</tr>
<tr>
<td>N</td>
<td>0.000477</td>
<td>0.192</td>
<td>1.581</td>
</tr>
<tr>
<td>O</td>
<td>0.000470</td>
<td>0.203</td>
<td>1.592</td>
</tr>
</tbody>
</table>

The effect of bubble size on the bubble-induced resistance were studied in the simulations of H, I and J. The bubble coverage was kept at a constant 50% with the bubble sizes at 65, 32.5 and 16.25mm. As the bubble coverage is the same on these simulations, the bubble gap is
similar to the bubble size. When the bubble size was increased, the predicted resistance increased from 0.000426Ω for 16.25mm bubble size to 0.00113Ω at 65mm (shown in Figure 7). These results show that the bubble size has a significant effect on the resistance at constant bubble coverage. The global current flow was found to be similar to that of the results in Figure 5 and Figure 6, the close-up images of the studies H, I and J are shown in Figure 8. They predict the local concentration of current density at the intersection point to increase with the size of the bubble [7].

Figure 7. Bubble induced resistance for the cases H, I and J [7]

Figure 8. Current density plot for cases H, I and J [7]
Zhang, et al. [7] also studied the bubble thickness with cases L, M, N and O. Where the bubble thicknesses were 3, 4, 6 and 7 mm, respectively, at a bubble coverage of 50% and 16.25 mm bubble size. The predicted results are shown in Figure 9 with case J. It can be clearly seen from the plot, with rising bubble thickness, the resistance due to bubble increased. According to Zhang, et al. [7], the increase in bubble resistance due to increase in thickness would be more significant at lower ACD. Moreover, the non-uniform current density vectors near the bubbles, will affect the motion of bubble by non-uniform Lorentz force induced in the bath. Bubble coalescence is a complex occurrence in an industrial cell that forms large bubbles. The case K has a bubble coverage at 100% with the bubble thickness at 5mm. The current density plot for the global current density is shown in Figure 10. It can be seen that the current flow avoids the underside of the anode (bubble layer) and flows from the side of the anode towards the central region of the metal. As a result of this by-pass, the bubble-induced resistance is approximately 50 times higher than other cases. Despite this significant change in current flow in the bath, the current flow in the cathode and collector bar domain has a relatively similar flow pattern to that of other cases [7].

![Figure 9. Bubble resistance for increasing bubble thickness at 50% coverage and 16.25mm bubble size [7]](image)

![Figure 10. Current density vector map with a bubble covering the entire underside of the anode (Case K) [7]](image)
The study of bubble-induced electrical resistance by Zhang, et al. [7] is valid only when the bubble is static with a fixed shape, however, the bubbles under a surface move due to the buoyancy and surrounding fluid motion induced by bubble and MHD induced flow. Therefore, the electrical resistance by bubbles may be different in a transient model. The importance of understanding bubble behaviour in the aluminium reduction cell is paramount for error free and efficient operation of the cell. The various factors and circumstances of bubble-induced resistance has been discussed to verify the importance of the role that bubbles play in terms of their electrical resistance. The removal of bubbles from the anode (gas holdup under the anode) contributes to less bubble induced resistance, better wetting of the anode, homogenisation of the bath concentration and temperature, and avoidance of the anode effect [4, 7, 10].

1.7.3 Bubble characteristic underneath a surface

The behaviour of bubbles underneath a surface is an important phenomenon that has been studied by many over the past few decades. The nucleation of a bubble under a horizontal surface is known to differ to that of bubble growing under a tilted surface. The formation is predominantly influenced by the gravity forces and the force balance between surface tension, viscous forces, buoyancy, gravity field and inertia. These are progressively changing with the growth of the bubble. The morphology of the bubble in the early stages of growth influences the onset of the bubble-induced flow field in the bath. For the growth by a nozzle, under a horizontal plane, the bubble initially expands while its base is attached to the nozzle [5]. The interface at equilibrium maintains a spherical shape governed by the well-known Laplace equation [38] given below.

\[
γ \left( \frac{1}{R_1} + \frac{1}{R_2} \right) = p^* + (\rho_l - \rho_g)gz
\]

Where the \( γ \) is the surface tension, while \( R_1 \) and \( R_2 \) are the first and second radius of curvature, respectively. \( p^* \) is the pressure drop across the interface at the tip of the bubble \((z = 0)\). The \( \rho_l \) and \( \rho_g \) are the density of liquid and gas, respectively, with \( z \) as the vertical height [38]. Figure 11 depicts the components of forces that act on a bubble growing under a horizontal surface. The force at the three-phase contact line due to the internal pressure of the bubble is given as \( F_p \). The bubble is attached to the surface by the forces given below:

\[
F_1 = 2\pi \cdot r (\sigma \sin \theta_c + F_p \cos \theta_c)
\]
Both $F_1$ and $F_2$ act upward causing the bubble to adhere to the surface, therefore the bubble is unable to detach in a perfectly horizontal plane. The horizontal component of surface tension force opposes the expansion of the bubble, while the force of the internal bubble pressure expands the base diameter. They can be expresses as $F_3$ and $F_4$ [5].

\[ F_3 = 2\pi r \sigma \cos \theta_c \]  \hspace{1cm} \text{eq. 20}
\[ F_4 = 2\pi r F_p \sin \theta_c \]  \hspace{1cm} \text{eq. 21}

The contact angle of the bubble changes rapidly in the initial formation period. As the $\theta_c$ trends towards 90° the force of $F_3$ reduces with rises in $F_4$, leading to the bubble shape changing from spherical to an elliptical shape. Similar to the aluminium reduction cells, the growing bubbles remain at their nucleation site until drag force by the velocity of the surrounding fluid overcomes the interfacial force [5, 39].

The bubble growth underneath inclined a surface undergo a sliding process where the detached bubble initially slides while maintaining a contact with the surface, which is known as the creeping zone as show in Figure 12(b). As the bubble continues to slide the nose lift’s off the surface as shown in Figure 12(c) to initiate the wetting film [40]. Aussillous & Quéré [41] also reported on the thin layer of lubricating film that forms between the bubble and the surface.
Figure 12. Stages of bubble movement as reported by Perron et al. [40]

The primary forces that effect a sliding bubble are the buoyancy force \( (F_b) \), surface tension and drag force \( (F_d) \) shown in Figure 13. The \( x \) and \( y \) components of the buoyancy force, flattens the bubble and initiates bubble sliding, respectively. The drag force opposes the motion of the bubble with the surface tension force influencing to sustain a spherical shape [42]. The surface tension can also affect the detached bubble size and frequency of the detaching bubbles [43]. Once the detached bubble has reached the terminal velocity, the two primary forces that influence the bubble are the buoyancy force and the drag force. These forces act in the opposing directions with similar magnitude, although on an inclined surface only the parallel component of the buoyancy force acts opposing the drag force [42].

![Figure 13. Illustration of the force balance on a gliding bubble [42](image)](image)

One of the earliest studies of rise of gas bubbles under an inclined surface in non-circular passage has been studied by Maneri & Zuber [44]. They utilized a two dimensional tank to investigate the effect of fluid properties and the effect of inclination on the rise of bubbles. The influence of the fluid properties was found to be negligible for vertically rising bubbles, while it markedly affected the velocity of bubbles rising in inclined planes. Clift, et al. [45] reported extensively on the motion of bubbles, droplets and particles. Maxworthy [3] investigated characteristics of bubble sliding in an large container where the effects of all walls except the sliding wall were negligible. The shape and velocity of bubbles were determined for surface inclinations of 5° to 90° with bubble volumes from 5 to 60 ml at 5 ml intervals. At
lower tilt angles with higher bubble volume the effect of gravitational flattening was more dominant. The bubble shape in such conditions were seen to resemble a boomerang shape, while at higher inclinations of nearly 90°, the resulting shape was more of the spherical cap. The velocity of the sliding bubbles were improved by the higher wall inclination and bubble volume due to the greater buoyancy force as shown in Figure 14. Moreover, both Xiang-peng, et al. [46] and Fortin, et al. [37] reported that, for a physical model with continuous generation of bubbles, the detached volume of bubble was found to reduce with increasing wall inclination. The effects of wall inclination on electrolytically nucleated bubbles were similar to that of air-injected bubbles. Thus, at higher surface inclinations the shorter bubble holdup time results in lower bubble coverage area, lesser bubble layer thickness and most importantly, lower bubble-induced resistance [6] (shown in Figure 15).

Figure 14. Bubble velocity for a range of bubble volume and surface tilt angles [3]
The nucleation of bubbles under an industrial anode results in coalescence of these bubbles into larger bubbles that move towards the ends of the anode. Fortin, et al. [37] designed a physical model that grows bubbles through a micro-porous polyethylene plate to form a large bubble with a thick nose and thin long tail that is commonly known as the Fortin bubble (shown in Figure 16). A number of other authors have also reported on similar bubble shape [47-50].

1.8 Magnetohydrodynamics (MHD)

The high electrical current (180-500 kA) that is required by the aluminium reduction cell generates an intense magnetic field within and around the cell [9, 51]. The majority of the magnetic field that is formed inside the reduction cell is formed by the electricity in the busbar system [20]. Typically, the current density in the external cathode bus, range from $10^2$ to $10^3$ kA/m$^2$, varying upon the size of the conductor. For example, a cathode bus of a 140 kA cell is shown in Figure 17. The current in the busbar near the duct-end and tap-end ranges from 105-108 kA, which in a 140 kA line is at 519 kA/m$^2$ that results in greater MHD forces near the endwalls of all end-riser cells [12]. The current density vectors ($J$) in the conducting liquids interact with the external magnetic fields ($B$) to give rise to volumetric forces called Lorentz force (eq. 22) which creates loops on the length scale of the cell itself [20, 22, 52].

\[
F = J \times B \quad \text{eq. 22}
\]

\[
F = JB \sin \theta \quad \text{eq. 23}
\]
Figure 17. Top view of the busbar and lining with current boundary conditions of a 140kA side riser cell [12]

The main source of instability in the metal pool is due to the horizontal component of the current density vector interacting with the vertical component of the magnetic field [53]. Das and Littlefair [52] conducted a numerical study to predict the current distribution and Lorentz force within an 300 kA end-riser aluminium reduction cell. As expected, the electricity travelled in the path of least resistance. Figure 18 shows that most of the current in the cathode is concentrated in the outer third of cathode. Figure 19 shows the current path lines from the anode to the cathode. The current flows vertically off the bottom surface of the anode, through the electrolyte to the metal pad. This is due to the high electrical resistance of the bath to that of the metal pad. The current then travels from the metal deposit to the cathode and then to the collector bar. The current has a large horizontal component (\(J_x\)) in the aluminium deposit than the cathode and travels almost vertically to the collector bar that is even less conductive [9, 52]. Thus, the more skewed (\(\theta\)) electrical path in the metal pad in comparison to the bath, induces stronger Lorentz forces (eq. 23) in the metal pool. This provides the rationale behind the greater Lorentz force predicted in this medium.

The induced forces set the aluminium deposit and bath in motion and deform the interface (heave) that would normally remain flat and horizontal when undisturbed [28]. The heave is the rise in the aluminium deposit from the flat horizontal plane due to the MHD forces, especially when the ACD is lower [12]. The MHD forces also induce a phenomenon called the metal pad rolling, which is the oscillation of the interface with a period ranging from five seconds up to more than a minute [54]. The gas bubbles evolving from an anode does not
experience the effects of the Lorentz force directly. However, the conducting fluid’s pressure, density, surface tension, viscosity and gravity field are considerably affected [1].

Figure 18. Vector map of the current density [52]

There have been several numerical models based on interface tracking methods that were reported in recent times. One of the most common method is to solve the magnetic diffusion and Lorentz force equations simultaneously, in order to obtain the magnetic flux density and the force field. Then the conservation of mass and energy equations are solved incorporating the force field as a source term sequentially. The governing equations including the Maxwell’s equations are given below [12].

\[
\nabla \times \vec{H} = \dot{\vec{j}} + \frac{\partial \vec{D}}{\partial t} \\
\nabla \times \vec{E} = \frac{\partial \vec{B}}{\partial t} \\
\nabla \times \vec{B} = 0 \\
\n\vec{j} = \sigma \vec{E}, \quad \vec{D} = \varepsilon \vec{E}, \quad \vec{B} = \mu \vec{H}
\]

eq. 24

eq. 25

eq. 26

eq. 27
The electrical current distribution is predicted by the Ohm’s law as follows [12]:

\[ J = \sigma (E + \vec{v} \times \vec{B}) \]  

eq. 28

The magnetic induction is determined as follows [12]:

\[ \frac{\partial \vec{B}}{\partial t} = \nabla \times (\vec{v} \times \vec{B}) + \frac{1}{\sigma \mu} \nabla^2 \vec{B} \]  

eq. 29

The term \( \nabla \times (\vec{v} \times \vec{B}) \) is negligible, in contrast, to other terms in the induction equation. The induction equation is decoupled from the velocity field, thus, the equation reduces to the following [12, 55]:

\[ \frac{\partial \vec{B}}{\partial t} = \frac{1}{\sigma \mu} \nabla^2 \vec{B} \]  

eq. 30

The continuity equation and Navier-Stokes with the two dimensional force field term are expressed as given below:

\[ \nabla \times \vec{v} = 0 \]  

eq. 31

\[ \frac{\partial \vec{v}}{\partial t} + (\vec{v} \times \nabla)\vec{v} - \nu \nabla^2 \vec{v} + \frac{1}{\rho} \nabla \rho = \frac{1}{\rho} \vec{F} \]  

eq. 32

Where the \( \vec{F} \) is the volumetric source term in N/m³.

A number of studies have been conducted to study the positive aspects of MHD forces in the cell, mainly as a source of bath momentum which improves the alumina transport and dissolution. Additionally, several studies have been conducted on achieving the stability in the bath-metal interface [12, 20, 56]. These studies aim to reduce the MHD forces in the metal pad in order to reduce the interface deformation and fluctuations, so that the ACD can be lowered further while avoiding short circuits. The positive aspect of lowering the distance between the anode and the metal is that it reduces the distance the current has to travel in the highly electrical resistive medium (bath).

Zhang, et al. [10] developed a CFD model for alumina-mixing process. The mixing process of alumina driven by bubbles and by MHD forces were calculated separately to assess their individual contributions to the mixing process. It was found that the alumina concentration for the bubble driven flow is highly symmetrical, in contrast, to the electromagnetic force (EMF)
driven flow where the alumina dispersion is strongly affected. The diffusion rate was found to be very low, however, the alumina was transported well by the entrainment effects of bath bulk flow as shown in Figure 20.

![Alumina concentration distribution on a horizontal view at 20s intervals](image)

Figure 20. Alumina concentration distribution on a horizontal view at 20s intervals (a) bubble driven flow (b) EMF driven flow [10]

The flow field (vertical plane) of the centre channel under the first alumina feeding point is shown in Figure 21. A horizontal motion of the bath is observed under the anode by the EMF induced motion, thus, alumina is transported to most locations of the cell regions. Figure 22 shows the contour of alumina distribution after 15 seconds of dispersion. The observation from these figures show that the bubble induced flow has a significant effect on the vertical motion on the initial stage of mixing, and without the bubble effect, the alumina concentrates on the top of the channel. The mean velocity of the upper channel in the bubble induced flow is 0.043 m/s and with EMF it is 0.017 m/s. The faster mixing of alumina in the local areas is expected due to the bubbles breaking up the agglomerated bulks of alumina, instead of transporting the alumina throughout the cell [10].
Zhang, et al. [10] concluded that EMF induced flow allows the alumina to be transported long range within the cell, while the bubbles increase the local alumina dispersion in the cell. Therefore, both forces have to be taken into account in modelling, as both factors play a positive role in uniform distribution and dispersion rate of added alumina. However, a few noteworthy assumptions have been made in this study. Firstly, the complexity of bubble behaviour in an actual cell including bubble to bubble interaction is not characterised in this CFD model. The bubbles are modelled as particles that have a given diameter without the coalescence. Secondly, the bath-metal interface motion has not been modelled, hence, a greater component of the Lorentz force in the cell is not present. The motion of the interface may contribute to the bath velocity, thus, different alumina mixing may be predicted.

Doheim, et al. [11] modelled a two-dimensional domain of an 208 kA aluminium reduction cell (Figure 23) to study the effects of bubble and MHD induced flow. The MHD forces were introduced as a source term in the governing equation. The motion of bubbles has been assumed to be unidirectional from the centre of the anode towards the channel width. The k-
The epsilon model also accounts for the wake-induced motion of gas bubbles. The velocities of the domain were calculated with bubble (Figure 24), only MHD (Figure 25) and both induced forces (Figure 26). The bath velocity under the anode region with bubble, EMF and combined forces, were found to be at 16.9, 5.8 and 25.2 cm/s, respectively, towards the projection of the anode. Whilst in the opposing direction, near the cathode, the velocity was at 3.8, 2.8 and 6.5 cm/s, respectively.

Upon comparison of the bubble and MHD induced forces, it is evident that the motion induced by bubbles are more significant than MHD induced flow in the modelled region. Moreover, the bubble induced flow also has a greater impact on the angular velocity. A similar angular velocity was present in the channel width region on the combined forces simulation [11]. The study also included the mass-transfer coefficients and current efficiency factors. Doheim, et al. [11] concluded that the bubble-induced flow is more effective in the bath in comparison to the MHD induced flow.

![Figure 23. Modelled domain (YZ) with all dimensions in centimetres [11]](image)

This study predicted that without the MHD forces the bath velocity under the anode was 33% lower than the CFD simulation with the combined forces. However, as the numerical model is only a two dimensional model, it would have lost the majority of the MHD forces as it is primarily significant in the third dimension. In addition, the bath-metal interface was not modelled where it may also contribute to the bath velocity. Therefore, the overall contribution by MHD forces to the bath velocity may be greater than the predicted results by this numerical model.
Das, et al. [9] examined the effect of steel and copper collector bars at various insertion lengths on the current distribution and induced Lorentz force in an 3D numerical model of one quarter of an industrial cell. Their aim was to reduce the MHD forces in the cell by improving the current distribution. The predicted force field with a steel collector bar at 100% insertion is at 160 N/m$^3$ and in the case of the copper collector bar, at 100% it is at 80 N/m$^3$. Figure 27 shows the contour plots for the Lorentz field in the molten metal. Despite the 50% reduction in maximum current density with a uniform current distribution, the drawbacks of the copper collector bars are low melting point, possible contact issues with the cathode and higher thermal conductivity (heat loss from the cell).
Figure 27. Molten metal contour plots (yz plane) of the Lorentz force (a) Steel collector bar at 100% length (b) Steel collector bar at 84% length (c) Steel collector bar at 72% length (d) Copper collector bar at 100% length (e) Copper collector bar at 84% length (f) Copper collector bar at 60% length [9]

Wang, et al. [20] developed a transient CFD model to understand the effects of the innovative cathode. The innovative cathode has rectangular protrusions (Figure 28) that is expected to reduce the inclined current path in the metal deposit (Figure 29), hence, the metal instabilities are expected to be reduced. A 3D model was used to assess the metal flow and interface fluctuations due to MHD forces and combined induced flow. In order to reduce computational time, a separate 2D model was used to study the metal flow and interface fluctuations due to bubble induced flow. The structured meshes of the 2D and 3D models were tested for grid independence and mesh with 350,000 and 1,700,000 elements were selected for the 2D and 3D models, respectively. The MHD forces were calculated on ANSYS and interpolated to the momentum equation by a user defined function (UDF) programme.

Figure 28. CAD model and image of (a) Traditional cathode (b) Innovative cathode [20]
Figure 29. Schematic representation of current path in the traditional and innovative cell [20]

As expected, all three components of velocity in the metal were lower in the innovative cell, for example, the average velocity of the metal in the traditional cell was 0.1142 m/s and with the innovative cathode cell it was reduced to 0.0754 m/s. Moreover, the interface deformation was higher in the traditional cell than that of the innovative cell. The bubble velocity at a monitor point in the top of the cell, between two cells is shown in Figure 30. The plot shows the velocity of the bubbles at this point for both types of cathodes are similar up to approximately 12.5 s and thereafter the bubble velocity is higher in the traditional cell [20]. Even though these results are encouraging, the bubble release study was conducted without the MHD forces in the metal pad and the bubbles are generated at given points under the anode, which is not similar to the electrolytically generated bubbles. In the case of the bubble velocity measurement, only one monitoring point was assessed in this study and the effect on the bubbles by reducing the MHD forces in the cell was not assessed in this study.

Figure 30. Bubble velocity for traditional and innovative cathode cells at one monitor point [20]
Figure 31. Interface amplitude plot for three points on both cells with MHD forces and bubbles [20]

The interface amplitude was also found to have been reduced by a considerable margin, as shown in Figure 31. Thus, altering the current distribution in the metal pad markedly affected the flow field induced by the MHD forces. Although, many authors have reported on the necessity of the MHD forces. Where they have been linked to many positive aspects of the cell process. The studies that aim to reduce MHD forces in the metal pad have not accounted for the loss in bath velocity and how they will affect the electrolysis process. A significant reduction of MHD forces in the cell can increase the bubble release time from the anode, thereby inducing higher bubble-induced resistance. In a more severe case, it may shift the current path from the underside of the anode to the sides. This further increases the distance that current have to travel from the anode to the metal pool.

Additionally, lowering the ACD can be problematic with the formation of large bubbles. It could limit the movement of the electrolyte to the bottom of the anode, which reduces the homogenisation of the bath temperature and electrolyte concentration. In addition, lowering the ACD can adversely affect the ability of the cell to generate its own. The waste energy generate from the current traversing in the bath contributes to self-heating of the cell (heat generating resistor) [24]. Which helps to maintain the cell temperature at optimal conditions without requiring an additional heating source.
1.9 Physical Modelling

The high temperature and corrosive bath of the aluminium reduction cell severely limits the study of generated bubbles [13]. Moreover, it is difficult to scale the Hall-Héroult process to laboratory scale. Thus, many have opted to developing and studying physical models that represent an actual cell, while maintaining some geometrical and dynamic similarity. In the study of bubble behaviour, the two primary types of physical models are the air-water model (also known as cold water model) and the electrolytic cell model. The most distinguishable differentiator between these two models are their method of generating bubbles. The air-water model injects air from under a wall, while the electrolytic cell model generates bubbles through an electrochemical reaction.

The most widely used physical modelling technique is the air-water model due to the ease of setup and operation. Additionally, the two systems, water and cryolite have similar kinematic viscosity [18, 57]. The ability to control the bubble growth site, injection rate and orifice size allowed the study of the bubble behaviour in a fundamental level. Additionally, the surrounding fluid properties can be altered while maintaining the same bubble formation parameters. For example, the addition of aqueous sodium dodecyl sulphate (SDS) to change surface tension [5].

There are several variations of the air-water model, the injection of air from an orifice underneath a surface [58], injecting air through a porous medium [37, 43], injecting air into an inverted cup that is then released [59] and injecting gas bubbles that rise into a wall [4, 60, 61]. Also, the simple mechanism of bubble generation has allowed many to develop CFD models that rely on the experimental data for validation and setup. For instance, Zhang, et al. [7] utilized an air-water model to obtain the bubble morphology, which was used to setup a numerical model that studied the bubble-induced electrical resistance. To attain geometric similarity, Feng, et al. [18] utilized a full scale three anode model (Figure 32) with anode dimensions of 1300 mm x 650 mm x 600 mm which represents an anode of a modern pre-bake smelter. The bath velocity was measured through PIV technique that was used to validate a time-averaged bubble driven flow numerical model.
The simplicity of the bubble generation process allows further modifications of this model. To achieve both dynamic similarity and geometric similarity Yan, et al. [62] modified the air-injection model (Figure 33 (b)) with safe and inexpensive fluids at room temperature. Water was used to simulate the liquid aluminium, whilst a light vegetable oil simulated the bath and the injected air simulated the anodic gas. Fundamentally, it simulates an (a) aluminium fluctuation layer region (b) electrolyte and gas bubble zone and (c) electrolyte transition layer as shown in Figure 33 (a). These fluids were chosen for their matching density and viscosity ratios as similarly as possible (see Table III). The aim was to utilize these two fluids to study the penetration depth and the rules of penetration depth.
Table III. Comparison of a modified cold water model with an industrial cell [62]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Model</th>
<th>Cell</th>
</tr>
</thead>
<tbody>
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<td>ρ&lt;sub&gt;n&lt;/sub&gt;, kg/m&lt;sup&gt;3&lt;/sup&gt;</td>
<td>0.9</td>
<td>2.1</td>
</tr>
<tr>
<td>ρ&lt;sub&gt;lt&lt;/sub&gt;, kg/m&lt;sup&gt;3&lt;/sup&gt;</td>
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<td>2.7</td>
<td>1.3</td>
</tr>
<tr>
<td>v&lt;sub&gt;in&lt;/sub&gt;, mm&lt;sup&gt;2&lt;/sup&gt;/s</td>
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</tr>
<tr>
<td>σ&lt;sub&gt;n-m&lt;/sub&gt;, N/m</td>
<td>0.03</td>
<td>0.55</td>
</tr>
<tr>
<td>σ&lt;sub&gt;n-η&lt;/sub&gt;, N/m</td>
<td>0.02</td>
<td>0.14</td>
</tr>
</tbody>
</table>

The air injection model is not an actual representation of the bubble formation mechanism in an aluminium reduction cell. Alam, et al. [6] stated two major issues with this type of physical modelling. Firstly, the bubbles that the air-water model generate have momentum (particularly injection by a nozzle). In contrast, bubbles that are generated electrolytically have no momentum at the point of nucleation and the nucleation sites depend on the chemical reaction. Secondly, the MHD forces play a vital role in the bath momentum, however, the air-water model does not simulate such forces. They concluded that the data that is collected by an air-water model is inapplicable directly to an actual aluminium reduction cell. They built a low temperature electrolytic model (Figure 34) that maintained complete geometric similarity (1/4<sup>th</sup> scale industrial cell) with five dimensionless numbers considered to maintain the dynamic similarity.

Qian, et al. [63] conducted a comparative study between the bubbles generated electrolytically (Figure 35) and by an air-water model (porous plate). However, the electrolytic cell generated bubbles at both electrodes, in contrast, to an industrial aluminium reduction cell where the bubbles are formed only at the anode. Alam, et al. [6] was able to avoid this issue by the use of a copper (II) sulphate solution as the electrolyte with 20% sulphuric acid. Although both electrolyte models were unable to generate MHD forces within the cell.
The current physical modelling techniques that have been used on the study of bubbles in a Hall-Héroult cell cannot replicate the bath with both bubble-induced flow field and MHD forces. The nucleation of bubbles under an anode with zero momentum at the point of nucleation is a characteristic of an actual industrial cell that the electrolyte model can simulate. The bulk motion of bubbles also represents an important factor that the electrolyte model is able to simulate. Despite this, the loss of MHD induced flow affects not only the bath velocity, but the effect on the bubble shape and its removal rate. It is noteworthy that the experiment by Yan, et al. [62] with the presence of two fluids with improved dynamic similarity was an improvement to the existing air-water model, however, the stated issues still persist in this type of physical modelling.

1.10 High Speed Photography

High speed photography is a well-known method of observing and studying bubble characteristics [5, 6, 62, 64, 65]. Their ease of use, image quality and acquisition rate has improved vastly over the years. The high speed photography method is not only used as a stand-alone method, but it is also a method of validating CFD models. Das, et al. [5] utilized high speed photography to validate a numerical simulation to study bubbles formed under a surface by air injection. Caboussat, et al. [48] employed videography, PIV and CFD techniques to study Fortin bubbles.

The captured images are processing and analysed by image processing software, such as, ImageJ[66] and GIMP (GNU Image Manipulation Programme)[67]. These tools can be used to clarify the captured data and extract further data. Alam, et al. [6] observed the bubble generation method, development, detachment and coalescence by the use of a high speed camera. They used a GIMP image processing software to correct an image distortion. The
bubble contact angle, diameter and thickness were measured shown in Figure 36. The measured bubble diameter was used to calculate the bubble coverage (\( \varphi \)) for the 100 x 150 mm\(^2\) area of the anode. The volume of bubble per anode area was also calculated with the determined bubble coverage and the bubble layer thickness. The high frame rate allows the images of bubbles to show the morphology of the bubble in detail and the transient (subtle) changes they undergo. For instance, a type of bubble that may be observed are Fortin bubbles [20, 48] and events of bubble coalescence.

![Figure 36. Side view of bubbles under anode with contact angle (\( \theta \)), length (l) and thickness (d) dimensions [6]](image)

The bubble behaviour in electrolytic cells are far more complex in nature than air-injected models. High speed videography is particularly important since numerical simulations are yet not able to model bubble behaviour in electrolytic cells. Capturing the bottom view of the anode can provide a number of information about the process, such as, the individual bubble path, coalescence process, bubble coverage, bubble size variation. Bubble tracking techniques can be utilized to tracked multiple selected bubbles and output their individual velocities for a series of image sequences. For example, MTrackJ plugin [68] for ImageJ is an add-on that tracks objects and produce their velocity and bubble-path.

1.11 Particle Image Velocimetry

The particle image velocimetry is a non-intrusive laser optical measurement technique that provides instantaneous full-fluid flow velocity vector measurements [69]. It enables the measurement of high-speed flow fields [70]. The PIV method has been used in many studies that focused on the effect of sliding bubbles, specifically, it has been a vital tool in the research of cold model setups that study air-water system in similar geometrical conditions to a Hall-Héroult cell [18, 48, 71, 72].

Cooksey and Yang [72] utilized the PIV method, primarily in evaluating key anode design parameters. For example, the presence of a longitudinal slot in a partial full-scaled air-water model of a Hall-Héroult cell. Furthermore, they acknowledge the suitability of the PIV technique in capturing and quantifying the local fluid velocities and turbulence. Later on,
Cooksey and Yang [71] further studied the slot configuration effect on the bubble induced flow and bubble behaviour using a PIV as the only source of data gathering. For instance, the effect of the slot width on the fluid flow is shown in Figure 37 with streamline plot and local turbulent kinetic energy contour superimposed on mean velocity vectors plot.

![Figure 37. PIV results on the slot width effect on the fluid flow in the centre channel [71]](image)

PIV measurements can be used as a secondary source of data and as a source of validation to a CFD modelling in studying the bubble induced flow [18, 48, 71, 73]. Feng, et al. [18] developed a CFD model of a full-scale air-water model of an aluminium reduction cell and as a source of validation they conducted a PIV study.

**1.11.1 Basic Principle**

The basic components of a PIV system consist of a light source (Laser) to generate a light sheet, a transparent container with seeded flow, suitable recording device and a computer with software to process captured images and generated velocity data. A light sheet is used to illuminate the particle seeded flow in the target area. The PIV system takes two images within a short delay and calculate the displacement of individual particles that have travelled within the time delay. The image is captured via a high quality lens on a single frame or on two distinct frames on Charged Coupled Device camera (CCD). The velocity is then calculated with the known time and the magnitude of displacement [70, 73, 74]. The overall process for a standard PIV is shown in Figure 38 from the Laser to the data analysis stage.
The standard PIV, also known as 2C-PIV, uses a single CCD or CMOS camera to determine two velocity components (in plane). The third velocity vector component is termed “invisible” for a standard PIV. However, this vector component can be obtained by other PIV methods, such as, the 3C-PIV (stereoscopic arrangement of two cameras) as shown in Figure 39, dual-plane PIV or by holographic recording [69, 70].

The captured images are divided into sections called interrogation areas (IA). These interrogation areas from both images are cross-correlated, pixel by pixel ($I_1$, $I_2$). A signal peak is produced by the correlation which detects the average particle displacement ($\Delta X$). Sub-pixel interpolation is used to achieve accurate displacement. This process is repeated for the whole target area to obtain the velocity vector map [75].
1.11.2 Seeding particles

The principle of PIV is based on determining the two fundamental components of velocity, the length and time. These measurements are determining the seeding particle velocity, thus, indirectly measuring the fluid velocity. This emphasises the importance of the fluid mechanical properties of the tracer particles to avoid substantial differences between the motion of the fluid and seeding particle [70]. The type of seeding particles used for a PIV depend on the fluid that is used. Generally, they must satisfy the three basic requirements where the particles should be able to reasonably follow the fluid flow without excessive slip and scatter adequate amount of light for the camera [75, 76]. Additionally, the seeding particles must not interact with each other, where they may also concentrate or disperse depending on the local flow conditions which may result in velocity bias errors [77-79]. Furthermore, the number of particles in the flow is quite important to gain a reasonable signal peak. Generally, 10-25 particles must be seen in each interrogation area [75].

The influence of the gravitational forces is a primary source of error if there is a mismatch between the fluid (ρ) and tracer particle densities (ρ_p). From Stoke’s drag law, the gravitationally induced velocity (U_g) can be derived. The particles are assumed to be spherical and in a viscous fluid at significantly low Reynolds number [70].

\[
U_g = d_p^2 \frac{\rho_p - \rho}{18\mu} g
\]

Where \(d_p\) is the particle diameter, \(\mu\) is the fluid’s dynamic viscosity and \(g\) is the gravitational acceleration. In a continuously accelerating fluid the velocity lag of a tracer particle can be estimated by the following equation [70]:

\[
U_s = U_p - U = d_p^2 \frac{\rho_p - \rho}{18\mu} a
\]

Where \(U_p\) is the velocity of the particle. The step response of \(U_p\) generally follows the exponential law if \(\rho_p > \rho\) [70].

\[
U_p(t) = U \left[1 - \exp\left(-\frac{t}{\tau_s}\right)\right]
\]

Thus, response or relaxation time is given as follows [70, 80]:

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\[ \tau_s = d_p^2 \frac{\rho_p}{18 \mu} \left( \approx \frac{d_p^3}{v} \right) \]  

eq. 36

Although, if the acceleration of the fluid is not constant or Stokes law cannot be applied the equations of tracer motion is more complex. Nonetheless, the relaxation time is suitable to determine the tendency of tracer particle to follow the flow. For different particle diameters in a strong deceleration in an air flow the response time is shown in Figure 40 by eq. 35. Thus, smaller diameter of particles ensures good tracking, however, the light scattering properties should be adequate [70, 76].

![Response time for oil particles with three particle diameters for a decelerating air flow](image)

Figure 40. Response time for oil particles with three particle diameters for a decelerating air flow [70]

The PIV technique has been frequently used in measuring flow fields of physical models that represent the aluminium reduction cells and verifying numerical models. The flow field information is vital, specifically, in the case of assessing the MHD as a source of momentum in the bath in comparison to the bubble induced flow. In the case of an electrolytic models, which cannot be numerically modelled, a PIV study can measure the flow field with relatively less complexity.

1.12 Summary and gaps in knowledge

It is clear that work reported in the literature, in the area of the bubble behaviour under inclined surfaces are limited to bubble induced hydrodynamics. Despite the renewed interest, the phenomenon of MHD forces in the electrolytic cell of an aluminium reduction cell have not been adequately studied as its importance to the operation of the cell is crucial. Several fundamental gaps of knowledge are found from the review of existing literature.
a. The introduction of a strong magnetic field on the characteristic of bubbles forming under a wall is not well understood. The change in the bath flow field can have drastic effects on the fundamental behaviour of the gas bubbles that ultimately influences the electrochemical reaction.

b. The majority of studies that have been conducted to understand the fluid flow in electrolytic cells do not account for the effect of MHD induced flow together with the bubble-induced flow. The lack of a physical model that can generate both flow fields have been the main reason for this limitation in knowledge. The debate on the dominance of MHD forces as a source of momentum in the bath exist.

1.12.1 Research question and scope

The present work aims to answer if the MHD forces play a dominant role in the bath flow field and how this affects the behaviour of the bubbles evolving underneath the anode. The high current flow in the conducting fluid within the cell together with the intense magnetic fields has been predicted to generate strong Lorentz forces. These forces are expected to significantly affect the behaviour of bubbles and improve the velocity bath flow field that may ultimately affect the electrolysis process. The abovementioned gaps in knowledge are addressed in this study by the following:

1. Modifying a low-temperature electrolytic physical model to investigate the impact of MHD forces on the evolution and sliding of electrolytically generated bubbles in an aqueous electrolytic medium and the effect of wall inclination.
2. Investigating the effect of MHD forces on a single gas bubble by a modified electrolytic model.
3. Studying the electrolyte flow field behaviour under bubble-induced, MHD induced and combined flow field (both sources of momentum).
Chapter 2

Influence of an External Magnetic Field on Electrolytic Bubbles

2.1 Introduction

The impact of gas bubbles nucleating underneath walls are applicable in many industrial applications. The significance of this process is vital to the efficient operation of the Aluminium reduction cells where the presence of gas bubbles play a key role in the cell process. The formation of gas bubbles underneath the anode have complex effects on the electrolysis process due to the presence of forces such as gravity, surface tension and inertia [1]. On one hand, the low electrical conductivity of bubbles increases the cell resistance for the traversing current, but on the other hand the hydrodynamics due to bubble-induced-flow homogenizes the alumina concentration and the temperature gradient in the bath.

Previous studies of the bubble resistance have been conducted using various physical modelling methods and they are categorised based on the mechanism of bubble generation. The majority of work in past decade have used physical analogous model (bench-scale) which generated gas bubbles by forcing air through a porous plate at room temperature. Experiments on these types of physical models (air injection into cell) may be suitable for understanding the impact of bubble resistance on the current flow but not the localised phenomena caused by the bubble shape, detachment and sliding mechanism. The air flow (mass inflow) into the cell would induce additional flow in the electrolyte/water (bath), which in turn influences the sliding characteristics of the bubble. However, in recent times, there have been renewed interest in bath-dynamics-research as many have successfully used electrolytically generated bubble in physical analogous cells. Despite the scaled-down electrolytic models that generate bubbles in similar principle to the Hall-Héroult cells [6], the impact of induced MHD forces within the bath have been largely neglected due to the inability of the scaled-down models to provide the complex current distribution that would interact with the magnetic field. The parallel arrangement of electrodes that have been used in these lab-scaled models engenders electricity to flow in a vertical manner in the aqueous electrolyte due to the poor electrical conductivity of the electrolyte. Thus, the resultant cross product of the current density vector and magnetic field has very little in electrolysis cell. In actual aluminium cells, current traverses in an inclined manner due to high electrical conductivity molten aluminium bath, which is main cause of MHD field in the cell. An inclined current path in the electrolyte is required to interact with the external magnetic field, which is not achievable in a geometrically...
scaled down model that uses an aqueous electrolyte with the traditional arrangement of parallel
electrodes. Therefore, this provides the motivation to develop and design a simplified model
with the ultimate goal to include the MHD forces within the aqueous electrolyte.

2.2 Experimental Setup and Methodology

In the aluminium electrolysis process, it is well known that the current traverses in an inclined
manner in the molten aluminium [12] due to high electrical conductivity of metal. The inclined
current path is the main source of MHD force in molten aluminium. However, it is impossible
to achieve a similar current distribution in a geometrically scaled-down model using aqueous
electrolyte. Thus, the experimental apparatus designed here aims to achieve some sort of
inclined current path in the electrolyte (dynamic similarity) rather than keeping the design
geosmetrically similar to the actual Hall-Héroult cell.

The electrolytic cell was made out of Perspex with dimension of 310 mm length x 160 mm
width x 173 mm height. Figure 41 shows the orientation of both electrodes in the electrolytic
cell. The cathode is positioned towards the far end of the wall, embedded onto the bottom
sidewall of the cell as shown the figure, to allow the current to traverse in a typical inclined
manner from anode to cathode. It is to be noted that the electrodes and electrolyte were chosen
carefully to avoid any bubble generation at the cathode. Thus, aqueous copper sulphate was
used as an electrolyte with stainless steel and copper as the anode and cathode, respectively.
The electrolysis process generates oxygen gas, which initiates the bubble nucleation on the
anode (steel anode). While the copper deposits on the cathode (copper cathode). The copper
electrowinning reaction (given in eq. 37) also generates sulphuric acid that in turn improves
the conductivity of the electrolyte.

\[
\text{CuSO}_4(aq) + 2e^- + H_2O(l) \rightarrow \text{Cu}(s) + H_2SO_4(aq) + \frac{1}{2}O_2(g)
\]

eq. 37

The steel-anode (75 mm length x 75 mm width x 5 mm thickness), and copper-cathode of (145
mm length x 40 mm width x 5 mm thickness) were prepared using fine-grain sandpaper and
rinsed with sulphuric acid and distilled water. The anode was insulated all around except the
bottom surface (thickness ≈ 0.5 mm), which allows bubble formation only on the downward
facing surface. The anode-holder-block was machined to hold the anode at different inclination
angles (θ ≈ 0°, 1° and 2° at ±0.05°).
Aqueous copper sulphate (CuSO₄) solution of 0.5 molar concentration was made up using anhydrous CuSO₄ in distilled water. The volume of electrolyte, molar concentration of aqueous CuSO₄ and the surface area of anode were calculated to inject 3 A of current into electrolyte using a DC power source. Which corresponds to an average current density of 800 A/m². It is to be noted that the current density in actual Hall-Héroult cell is much higher.

![Experimental setup with dimensions in mm](image)

Figure 41. Experimental setup with dimensions in mm

An insulating partition (barrier) with a rectangular slit was placed in between anode and cathode to provide a restricted path for the current flow. Figure 42 shows the rectangular slit on the bottom of the sheet, which also restricts the copper-deposits on the cathode to remain in side channel, i.e. in-between the insulating partition and the cell sidewall. The rectangular opening allows for the current to traverse from the anode to the cathode, in an inclined path.

A DC power supply (Instek GPC-3030DQ) was used to provide the current at constant rate of 3 amperes (800 A/m²). It is to be noted that it was difficult to maintain a steady current flow as the ionic concentration decreases with the electrolysis process. In the past, lab-based aqueous electrolytic cells have used a constant flow of electrolyte into the cell [6] to maintain the steady current flow. However, the inflow of fresh electrolyte into the cell would change the hydrodynamics of the electrolyte (surrounding fluid of the bubble), which in turn may have some influence on the bubble sliding characteristics.
A neodymium-based magnet (NdFeB) with a surface field of 1.8 T (Tesla) was used as the source of the static external magnetic field as shown in Figure 41. It is known that the magnetic field decays rapidly (exponentially) with distance from the magnet. Magnetic field strength was measured using a gauss meter which varies from 0.1 T (at the far end of the anode) to 0.72 T (approximately) at the sidewall. Figure 43 shows the variation of magnetic field (Gauss) from magnet-surface with distance.

A Nikon D7000 DSLR camera was used to capture the bubble evolution and sliding underneath the anode at 24 frames per second (fps). The DSLR camera was chosen specifically for its ability to capture images at Full High Definition resolution of 1920 × 1080 over a high image acquisition rate. In order to obtain an image with even lighting sheet, a diffuser is placed.
in front of the artificial lighting. The images were analysed in ImageJ, an image processing software. A manual tracking add-on (MTrackJ [68]) was used to measure the bubbles sliding velocities and their sliding-paths. The images were converted to 8-bit images to clearly identify the boundaries of individual bubbles. The images were calibrated with a length scale on the anode. The bubble velocities were determined by tracking the centroid of the bubble based on eq. 38. The bubble’s axial position was tracked from \( x_1, y_1 \) to \( x_2, y_2 \) at a time step of \((\Delta t=1/24\text{sec})\). Where \( K_p \), is the distance between two adjacent pixels with a unit of mm/pixel.

\[
v_b = K_p \cdot \frac{\sqrt{(x_2 - x_1)^2 + (y_2 - y_1)^2}}{\Delta t}
\]

eq. 38

2.3 Results and Discussion

Gas bubbles are observed to nucleate 0.05s after initiating the electrolysis process. The nucleating sites are seen to develop along the leading edge of the anode and outwards. The leading edge of the anode is the region with the shortest ACD. As the traversing current in the electrolyte takes the least resistance path, the leading edge of the anode is expected to have maximum current density. Figure 44 shows the images at 10 second time intervals, \((\Delta t = 10 \text{ s})\), for the horizontal anode \((\theta = 0^\circ)\) with no external magnetic field. The bubbles nucleate covering nearly the entire anode surface, which is observed as a formation of a thin gas layer transforming into bubble nucleating sites. The number of bubbles and their sizes increase rapidly with time in the region with relatively higher current density, i.e. the edge of the anode near to the side channel. As the bubble coverage area increases, the current density keeps changing in other parts of the anode. It was difficult to estimate the rate of bubble nucleation at a particular region of anode. Alam, et al. [81] also reported similar size of bubbles underneath the anode surface using copper sulphate electrolyte.

![Figure 44. Bubble motion at \( \theta = 0^\circ \) with no magnetic field](image-url)
Figure 45 and 46 shows the bubble motion underneath the inclined anodes ($\theta \approx 1^\circ$ and $2^\circ$) without any magnetic field. The bath flow is mainly caused by swarm of bubbles rising on the surface. The smaller bubbles gradually join to form larger bubbles due to coalescence process (Laplace pressure difference). The evolving bubbles were observed to slide away from their site of nucleation when the buoyancy force overcomes the interfacial force. This promotes localised phenomena such as wake by the electrolyte and/or more sites of nucleation. The enhancement of further gas nucleation signifies more mass transfer in electrochemical process. The smaller bubbles in a bubble swarm was observed to exhibit a sluggish flow at most parts of the anode.

![Figure 45. Bubble motion at $\theta \approx 1^\circ$ with no magnetic field](image1)

![Figure 46. Bubble motion at $\theta \approx 2^\circ$ with no magnetic field](image2)
Figure 47 and 48 shows the onset of MHD convection, from the bubble motion, in all parts of anode surface. A higher bubble sliding velocity was observed throughout the anode. The bubbles near the edge of anode have the highest velocity at highest magnetic field of 0.6 T. It can be seen that the direction of the bubble drifting path (white line on Figure 47 - 49) remains primarily parallel to anode edge as the Lorentz forces act orthogonally to both the current flow and magnetic field. This observation is noteworthy as the bubble sliding paths that were observed under the influence of a magnetic field varies markedly from the bubble-induced flows. The induced Lorentz forces swept most of the gas bubbles away from the anode edge.

![Figure 47. Bubble motion with magnetic field at $\theta \approx 1^\circ$](image1)

![Figure 48. Bubble motion with magnetic field at $\theta \approx 2^\circ$](image2)
Three distinct regions (zones) of bubbles under the anode can be observed: (1) bubble flow along the edge of the anode, (2) a swarm of bubbles and (3) few scattered bubbles growing in the inner regions of the anode. Figure 49 shows individual bubbles tracked in the bulk bubble region (zone 2) from the image sequence of time 16 to 27 seconds in the lower right hand corner of the anode as shown in Figure 45. The bubble paths are tracked by contrast measurement based on a maximum intensity technique. The drifting bubbles move linearly in the bubble-induced flow field. However, in the presence of the magnetic field, the swarm of bubbles are seen to form a rotational path due to the opposing flow field near the edge (zone 1). The anode with higher inclination results in more condensed swarm with shorter mean distance between bubbles. As a result, the behaviour of electrolytic bubbles is considerably affected by the characteristics of the swarm of bubbles and the bubble to bubble interaction. The bubbles that are been swept along the edge tend to coalesce with bubbles that are moving towards the edge.

It is now clear that bubble move in a cluster and exhibits a bubble swarm characteristics. For the horizontal anode ($\theta \approx 0^\circ$), it was difficult to track the velocity of any individual bubble due to very low velocities of these bubble clusters. Thus, velocity of the bubbles at the horizontal inclination was not considered for any comparison. Figure 50 shows the bubble average velocity vs. time graph for inclined anode. The velocity of the bubbles at location 9–10 mm away from the edge were calculated after 18 seconds of electrolysis process using the image processing software. Average velocity were calculated by considering a number of larger bubbles. The figure shows a complex velocity pattern as the bubbles slide/drift in a cluster or group of clusters. As expected the higher tilt angle of the anode improves the bubble velocity. The bubble sizes grow as they move towards the side channel. The velocity also increases gradually as the gravity begins to dominate for larger size bubbles. It was also found that the velocity of these individual larger size bubble increases rapidly in the side channel, i.e. in the vicinity of external magnetic field. Clearly, this shows the influence of the MHD field on bubble velocity.
Figure 49. Bubbles tracked for $\theta \approx 1^\circ$ in $T=16-27s$ (a) no magnetic field (b) with magnetic field

The average velocity of bubbles in zone 2, where bubble exhibits swarm characteristics, are found to be 1.12mm/s and 1.38mm/s for $\theta \approx 1^\circ$ and $2^\circ$, respectively. However, the bubble flow along the edge of anode (zone 1), exhibits a distinct movement (strongly parallel to the anode) due to the dominance of MHD field. Bubbles in this region of the anode have a notably higher velocity and found to be as high as 28.9 mm/s (Figure 51). The figure also show that the magnitude of these velocities are higher at higher inclinations.
The third zone, which is far end of the anode, shows few scattered and isolated bubbles due to lower current density at the anode surface. These bubble were found to be notably smaller in diameter to be influenced by any gravity field. After nucleation they tend to remain in the same location while growing at a very low rate. However, under the influence of the magnetic field, these bubbles were found to move in a rotational path (see Figure 52) towards the bulk bubble region while more nucleation sites were seen to evolve. The Figure 53 and Figure 54 shows the velocity of bubbles that nucleates at the far end of the anode (zone 3). The figure shows that the angle of inclination (from 1° to 2°) has negligible influence. The sliding velocity of gas bubbles in all regions of the anode were found to be enhanced in the presence of MHD field irrespective of the anode inclination. Matsushima, et al. [82] reported similar observation for the MHD field on bubble rise in water electrolysis with vertical electrodes. In general, the higher bath velocity in presence of a magnetic field not only improves the removal of bubbles off the conductive surface, but also reduces the bubble induced bath resistance [83]. Reduction in bath resistance has been reported to have a significant impact on the cell-efficiency due to reduced cell potential [84].
Figure 53. Bubble velocity in zone 3 at $\theta \approx 1^\circ$

Figure 54. Bubble velocity in zone 3 at $\theta \approx 2^\circ$
2.4 Conclusion

A laboratory based electrolytic cell was developed specifically to achieve dynamic similarity between the physical model and Aluminium reduction cell to investigate the impact of MHD induced convection. Typically, the dominant forces found to be governing the bubble behaviour are the inertial forces derived from the flow and the buoyancy force of the gas bubbles under bubble-induced flow condition.

However, the introduction of an external magnetic field during the electrolysis of aqueous CuSO₄, regardless of anode tilt angle, considerably effects the number of evolving gas bubbles, coalescence, sliding velocity and the bubble moving path. The enhancement of the coalescence process and the sliding velocity under a magnetic field is noteworthy, as it indirectly indicates the dominance of Lorentz forces in the bath over a simply bubble-induced flow field.
Chapter 3

Sliding Bubble in an External Magnetic Field

3.1 Introduction

The effects of MHD induced convection on electrolytically generated bubbles has been presented in previous chapter. The experimental data shows a strong interplay between the external magnetic field and the bubble velocities near side channel, i.e. in the region with higher current density. The experiment clearly demonstrates how bubbles nucleate instantaneously at the onset of electrolysis and how they grow with time. The electrolyte flow underneath the anode is mostly caused by swarm of bubbles rising on the surface. It is apparent that the coalescence process makes the bubble larger and begins to slide faster under the influence of gravity force. The enhancement of the coalescence process and the bubble velocity in the presence of a magnetic field is found to be notable, since it implies the dominance of MHD induced flow in the electrolyte rather than simply a bubble induced flow, as was previously thought. However, the main drawback of the experiment was the average bubble-bubble distance, which became shorter with increasing nucleating sites. The majority of bubbles, always remain in a cluster at any instant of time and its hydrodynamics (sliding behaviour) mostly depends on the characteristics of bubble swarm and consequently the bubble-bubble interaction. It was difficult to locate any single bubble evolving through all different stages of formation, detachment and sliding underneath the anode surface. Measurements of the shape and velocity of an individual bubble are rather difficult and may be practically impossible in the electrolytic process due to its inherent nature of nucleation process.

In this chapter, the aqueous electrolytic cell is modified to allow a single bubble to slide underneath the anode surface in a similar electrolytic condition to the Chapter 2 to study the influence of the external static magnetic field at a fundamental level. Most of the earlier experimental work, which reported the characteristics of a sliding bubble, use water as a surrounding fluid and uses a single air bubble. Furthermore, many published studies on the bubble behaviour on aluminium reduction cells have implemented the air injection technique to generate and study bubbles and their resulting flow field. Therefore, this chapter will provide the fundamental insight into the effect of MHD induced convection on a sliding bubble under a conductive surface.
3.2 Methodology

In order to achieve the inclined current distribution in the electrolyte whilst avoiding the generation of gas bubbles at either electrode some (minor) changes were implemented to the previous physical model. Copper was used as both the anode and cathode in aqueous copper (II) sulphate solution. The copper anode is preferentially oxidised to discharge $Cu^{2+}$ as both sulphate and hydroxide ions relatively more stable. The copper deposited on the cathode is equal to the copper dissolved at the anode. At the cathode these ions are reduced and deposited as given in the half equations below:

$$Cu - 2e \rightarrow Cu^{2+} \quad \text{eq. 39}$$
$$Cu^{2+} + 2e \rightarrow Cu \quad \text{eq. 40}$$

Figure 55 shows the schematic diagram of the revised electrolytic cell. Both the anode and cathode were prepared using fine-grain sandpaper and rinsed with sulphuric acid and distilled water. The anode was insulated all around, except the bottom surface, which allows the electrolysis to occur under the anode. The lack of gas generation at either electrode provides a bubble free electrolytic media. The bubbles are formed continuously by the injecting air through an orifice with a diameter of 3 mm. Two different orifice locations are studied as shown in Figure 56. The far end location (orifice 1) was used to analyse the sliding characteristics of bubble with and without the external magnetic field. The second orifice was chosen at the region with maximum expected current density on anode surface, i.e. near to the side channel. The copper cathode is embedded onto the bottom sidewall of the cell, similar to previous experiment, to create a current path in an inclined manner. It is expected that the MHD force would have the most dominating effect in the side channel due to higher current density and the direction of the traversing current.

A precision syringe pump (PHD Ultra, Harvard Scientific, USA) was used to inject air at a controlled rate of 6.2 ml/min into the quiescent electrolytic media. A Phantom V711 high speed camera was used to capture the bubble behaviour from the bottom view of the anode. The images were captured at 100 fps in 1020 x 800 resolution with an exposure of 3,000 $\mu$s. In order to maximise the contrast, a light source was placed behind the camera. An even light sheet was formed by the use of a diffuser. ImageJ, an image processing software, together with a manual tracking add-on “MtrackJ” [68] based on maximum pixel intensity was used to measure the bubble sliding/drifting velocity and their trajectories. The Images were converted to binary and a calibration marker (length scale) was used to scale the image using its pixel
length information. The bubble velocity was determined by tracking the centroid of the bubble using the eq. 38.

Experiments were conducted for three different inclinations ($\theta = 4^\circ$, $6^\circ$ and $8^\circ$ at $\pm 0.05^\circ$) of anode. The anode holders were made out of plastic to prevent any possible reaction with aqueous electrolyte. These anode holders were designed to keep the tip (leading edge) of the anodes at the same height, thus providing a similar magnetic field gradient across the anode plates. A neodymium-based magnet (NdFeB) was placed near the sidewall of the cell to provide the static external magnetic field.

Figure 55. Schematic of Apparatus (in mm)
3.3 Results and Discussion

3.3.1 Nucleating Bubble

It is well known that bubbles evolve and slide in an asymmetric manner along the vertical plane due to the buoyancy and surface tension forces resulting in distinct contact angles at both advancing and receding edges, however, they maintain a symmetric shape along the inclined (horizontal) plane when viewed from the bottom of the tank. Bubbles evolve in a spherical shape symmetrical to the axis of injection for all inclinations considered here. Figure 57 shows the images of a bubble evolving under a 4° inclined surface when injected at the orifice 2 (near leading edge) without the magnetic field. At the initial stage, the shape appears to be spherical due to the high pressure when the radius of curvature is small. It is believed that the shape is mostly governed by well-known Laplace equation that makes a radial outward growth during initial evolving phase. The gradual transition from spherical to a teardrop shape is seen in all inclination angles, which is attributed to mainly the gravity field. The buoyance force (the component parallel to the inclined plane) is the main cause of the bubble stretching along the inclined plane while the perpendicular component flattens the shape of the bubble. The stretching of the bubble results in larger radius of curvature, which initiates the necking and subsequently a detachment from the nucleating site. The symmetric shape during initial
evolution phase, as seen in Figure 57, are observed in both locations (orifice 1 and 2) without the presence of any external magnetic field.

Figure 58 shows the images of the evolving bubble in the presence of an external magnetic field at orifice 2. It is interesting to see that the shape, size and the stretching ratio of the bubble has change significantly by the MHD field created by the interaction between the cell current and the external static magnetic field. The change in the bubble orientation is initiated at the onset of nucleation. The bubble axis is found to be inclined approximately 24° towards the right side of the cell. This clearly demonstrates the influence of magnetic field from the onset of the nucleation. However, at location one (orifice 1), the evolving bubble remains unaffected showing a symmetrical shape even in the presence of the magnetic field. This is attributed to the decay of the magnetic field with distance, which was measured to be approximately 98% less at the orifice 1 (∼17.3 Gauss) than the anode edge (∼876.5 Gauss). Additionally, a low current density was expected near orifice 1 due to the larger ACD at this point.

![Figure 57. Bubble growth at time 0.05s time interval](image)

![Figure 58. Bubble growth with magnetic field at 0.05s time interval](image)
3.3.2 Bubble detachment

The sliding bubble behaviour underneath an inclined plane goes through three distinct phases: the bubble growth, detachment, and sliding along the inclined plane. So far, our current understanding on the bubble dynamics underneath a downward facing plate, indicates the dominance of buoyancy effect on the bubble detachment process from the orifice tip. Figure 59 (a) and (b) shows the sequence of images at 0.01s time intervals without and with the magnetic field, respectively ($\theta = 4^\circ$). It can be seen from the images that the bubble has a round nose with circular arc at the leading edge and a straight flatten body that joins to the tail. The figures clearly show the necking effect, i.e. the narrowing of the tail as the bubble gradually elongate. The neck begins to retract right after the detachment, but it is interesting to visualise the shape of bubble and their symmetricity during the detachment process in both cases, with and without magnetic field.

It is apparent from the figures that bubble volume is lower when detached in presence of external magnetic field. The bubble detaches approximately 2 seconds earlier in the presence of external magnetic field for 4° degree inclination plate reducing the bubble holdup time. The bubble holdup time is an important parameter in Hall-Héroult process as it changes the cell resistance, thereby the anode current density. The MHD field due to the external magnetic field not only initiates an early detachment from the orifice tip but also give rise an asymmetric trajectory after the detachment. Most of the numerical work that have been published suggest that the characteristics of bubble induced flow in Hall-Héroult is attributed to the symmetric wake behind the bubble. However, under the influence of an external magnetic field, the bubble sets out in an asymmetric trajectory after the detachment process.

These qualitative observations may relate to the detailed theoretical understanding of magnetic interaction parameter [1], which may be explained through such complex shape change process during combined rotation and translation motion of the sliding bubbles. The effect of MHD forces on a rising bubble was reported by Shibasaki et al. [85] in a liquid inside a rectangular enclosure by a 3D CFD model. They also found that with increase in the strength of the magnetic field (Hartmann numbers 0 to 200) the morphology of the bubble tend to elongate.
Figure 59. Bubble images (Δt = 0.01s) during detachment (a) without magnetic and (b) with magnetic field at θ = 4°
3.3.3 Sliding bubble

In this section the sliding characteristics of the bubble from the experimental model ($\theta = 4^\circ$, $6^\circ$ and $8^\circ$) over the entire length of anode are presented. The bubble at orifice 1 grew until it was sufficiently large enough to overcome the surface tension forces and slid in a nearly linear path towards the higher end of the anode due to the buoyancy effect as shown in Figure 60 (a). The air injection rate (6.2 ml/min) was selected to be sufficiently low enough to only allow one bubble to slide underneath the anode at one instance for all inclinations. The intention was to visualise and compare the sliding characteristics of an individual bubble in an electrolytic media with magnetic and without magnetic field. Figure 60 (b) shows the trajectory of the bubble in presence of an external magnetic field. The bubble tends to deviate gradually from its linear trajectory due to increasing MHD gradient towards the sidewall of the cell. The maximum deviation of the bubble occurs closer to the end of the anode.

![Figure 60. $\theta = 4^\circ$ inclination with the bubble trajectory (blue line) (a) without a magnetic field (b) with a magnetic field](image)

The gradual directional change of the drifting bubble (skewed towards the right) supports the existence of induced Lorentz force based on the orientation of the magnetic field vectors and the current density vectors as per the right hand rule. The electrolysis process of CuSO$_4$ with copper electrodes result in the transfer of copper from the anode to the cathode. The leading edge of the anode (shown in Figure 61) deteriorates at a higher rate than the other regions of the anode. This is expected since the current density tends to concentrate at the location of the
shortest ACD, hence, the uneven erosion. Therefore, this supports rationale behind the greater deviation of the bubble as it moves close to the end of the anode.

The continuous erosion of the anode changes the surface quality with time, which may affect the behaviour of the sliding bubble. The erosion can be seen to be biased in the direction of the bubble drifting path (right edge). This may be due to the sliding bubble’s wake that promotes the electrochemical reaction with fresh reagent. Similarly, the erosion of the anode is seen in the industrial aluminium reduction cells, where the carbon anodes participate in the electrochemical reaction. Although, due to the injection of bubbles instead of nucleating bubbles, the effect of surface deterioration is only applicable in a fundamental level to an industrial cell. Alternatively, in the previous study with steel anode, the bubbles were generated electrolytically, but the electrodes were non-participating in the electrolysis process, and thus, no electrode erosion.

To further clarify and substantiate this study, the orientation of the magnet was reversed from South to North facing upwards while maintaining all other experimental variables. This changed the direction of the predicted MHD forces towards the left, based on the right hand rule (see Figure 62). The change in Lorentz forces is expected to reverse direction of the flow field in the cell, thus, influencing the bubble sliding path. The Figure 63 shows the bubble path taken for the 8° inclination with both magnetic field orientations. In both conditions, the bubble escapes the surface with a similar x-axis deviation of 8.5 mm (left) and 8.0 mm (right) from the nozzle. Furthermore, the bubble flow path shows a consistent trend, with the bubble moving in the reverse direction and pivoting steeply in the latter half of the sliding process.
(0.3 s after detachment) on both conditions of the magnetic field direction regardless of their change in flow direction.

![Lorentz Force](image1.png)

**Figure 62.** Right hand rule for (a) North upward (b) South upward

![Bubble sliding path](image2.png)

**Figure 63.** Bubble sliding path at $\theta = 8^\circ$ for (a) North side and (b) South side magnet

The measured velocity of these two bubbles (show in Figure 64) show both velocity profiles are similar in trend and magnitude. Particularly, the higher peak velocity and loss of velocity until 0.5 s to a similar magnitude of the non-MHD experiment. The similar velocity and bubble path is engendered despite the directional change of the bubble drift as the polarity of the magnet is change and not the magnitude of the magnetic field strength nor the current density in the cell. Therefore, we quantify and establish that the forces that are introduced in the electrolytic cell by the external magnetic field are Lorentz forces and these forces are responsible for the skewing of the bubble path.
3.3.4 Effect of MHD forces on Bubble Creeping Morphology

Figure 65 to 65 show the representative sliding bubble sequence for all inclinations with their magnetic experiment counterpart. Each of these figures are a collage built from a sequence of images of the moving bubbles. After detachment, the bubbles undergo two stages of bubble morphology change from ellipsoidal to a shape with a wider nose and thinner tail. However, under the influence of the MHD forces this process is considerably affected.

For $\theta = 4^\circ$, Figure 65(a) shows the features of the single bubble at 0.1 s interval. At 0.3 s the bubble changes to an ellipsoidal shape and by 0.45 s the bubble’s nose is wider than the tail. Under the effects of the external magnetic field shown in Figure 65(b), the ellipsoidal shape is reached at 0.3 s. No major change in the bubble shape is seen until 0.40 s after detachment, but the orientation of the bubble is rotated towards the right. At 0.45 s, the bubble deforms unevenly under the stronger influence of the flow field.

Figure 66(a) shows a representative sequence for the $\theta = 6^\circ$ where the bubble shape changes to an ellipsoidal shape after 0.2 s. At 0.3 s the bubble undergoes the shape transition with thinning tail and wider nose. On Figure 66 (b) under the effect of MHD forces, the bubble changes shape to the ellipsoidal shape at 0.2 s, while the effects of the combined flow field results in bubble orientation shift at 0.3 s with notable shape change seen at 0.35 s.
For \( \theta = 8^\circ \) shown in Figure 67 the bubble shape change to ellipsoidal shape occurs more rapidly at 0.15 s and 0.1 s for bubble-induced and combined flow field, respectively. At 0.25 s the bubble undergoes the shape transition with thinning tail and wider nose. Under the magnetic field, the bubble path change is seen just after detachment at 0.1 s with shape change seen as early as 0.2 s.

A trend can be established where the shape changes are seen to occur at a shorter time span with higher inclination. The time difference between the shape changes from \( \theta = 4^\circ \) to \( 6^\circ \) is greater than the \( \theta = 6^\circ \) to \( 8^\circ \) change. Under the magnetic field influence, bubble transition to ellipsoidal for \( \theta = 4^\circ \) and \( 6^\circ \) as similar to the magnetic field experiment, although, at \( \theta = 8^\circ \) the shape change occurs 0.05 s quicker. Under the magnetic field, the deviation and major shape changes are seen rapidly with higher inclination. In contrast, no detectable changes were seen with the electrolytic bubble experiment in the previous chapter mainly due to the difficulty of the smaller bubble size and bubble coalescence.

![Image collage](a) (b)

Figure 65. Bubble path in sequence for \( \theta = 4^\circ \) anode inclination (a) without and (b) with a magnetic field (Image collage)
Figure 66. Bubble path in sequence for $\theta = 6^\circ$ anode inclination (a) without and (b) with a magnetic field (Image collage)

Figure 67. Bubble path in sequence for $\theta = 8^\circ$ anode inclination (a) without and (b) with a magnetic field (Image collage)
3.3.5 Comparison of Bubble Velocity and Sliding Path (without magnetic field)

Figure 68 shows a comparison of bubble sliding velocity underneath the plate at different inclination angles ($\theta = 4^\circ, 6^\circ$ and $8^\circ$). In general, the higher inclination of the plate result in smaller volume for the detached bubble and higher sliding velocity along the inclined path. The tail moves instantaneously after the detachment increasing the bubble velocity significantly immediately after the detachment. In general, the tail moves, in a way, to reduce the stretching ratio of the bubble and tends to attend a fixed stable length with further sliding. This type of behaviour results in non-uniform shape of the bubble during the time of detachment. The bubble begins to slides with a constant velocity after reaching a stable fixed length.

![Figure 68. Bubble velocity for non-magnetic experiment at 4°, 6° & 8° inclinations](image)

3.3.6 Comparison of Bubble Velocity and Sliding Path (with magnetic field)

For the experiments with the magnetic field, prior to commencing the electrolysis of the experiment with the magnetic field, bubbles were continuously injected, thus, the bath flow field was predominantly a bubble-induced flow field. When the electrolysis process is started with the magnetic field, the two fundamentally different sources of momentum interact and gradually form a consistent combined flow field in the bath. A gradual change in the flow field is seen with deviation of the bubble growing with time until a stable electrolyte flow is established. Once a consistent bubble path was reached, the bubble is tracked for analysis and comparison. Due to the two different mechanisms of bubble formation, the flow field of the
bath, between the electrolytically generated bubbles and the injected bubbles differ considerably. For the electrolytic bubble experiments in chapter 2, the bubble-induced flow field and MHD forces are initiated at the same time since both require the flow of electricity through the cell. Also other factors of electrolytic bubbles, such as, bubble to bubble interaction, no momentum at nucleating site, and nucleation site concentration near the edge would result to a considerably different bath flow.

Figure 69 shows a representation of bubble path along the inclined plane ($\theta = 4^\circ$). Figure 70 plots the deviation of the bubbles along the x-direction with time. The bubble path that has been tracked in ImageJ software, clearly illustrates the impact of the magnetic MHD field on its sliding trajectory. For $\theta = 4^\circ$, a gradual change in bubble path (towards the right) is seen with a maximum positive deviation of 17 mm from the centre axis (y-axis as shown in the figure). The increase in inclination results in smaller bubbles which exhibit a complex-convoluted motion in presence of the external magnetic field. This may be due to the combined effect of lesser gravity field on the bubble and the MHD field. It is to be noted that in the case of electrolytic process (chapter 2), bubbles usually evolve in the region of high current density, which experiences higher Lorentz field near the sidewall. Thus, experience a consistent bubble deviation from the onset of electrolysis.
The velocity of each individual bubbles were tracked and plotted against time for all inclination angles, with and without magnetic field. Figure 71 (a), (b) and (c) shows the comparison of bubble velocity at $\theta = 4^\circ$, $6^\circ$ and $8^\circ$, respectively. The characteristics of velocity pattern, i.e. a steep rise in velocity during detachment and then gradually attending to a constant velocity magnitude is a trend that is visible in all the examined cases. Although, in general, the presence of the magnetic field can be seen to enhance the velocity of the sliding bubble irrespective of the anode tilt angle.

The enhancement of bubble velocity in terms of its magnitude, seems to be not significant in comparison to the change in bubble trajectory (i.e. the deviation of sliding path) and the deformation of the bubble shape with time. Which would have a more substantial impact on the electrolysis process. Takatani [86] also predicted that a vertical magnetic field (perpendicular to the direction of gravity) deforms the rising bubble into more a complex shape, although the rising velocity was not affected greatly.
Figure 71. Bubble velocity (a) $\theta = 4^\circ$ (b) $\theta = 6^\circ$ (c) $\theta = 8^\circ$
In the absence of the external magnetic field, the bubble displacement is chiefly in the y-axis, hence, the y-component of the overall bubble velocity is dominant. The x-axis movement of the bubble in the non-magnetic experiments is low enough to be negligible. The induced Lorentz forces are expected to be primarily in the x-axis. The influence by the MHD forces in deviating the bubble path in the x-axis may hamper the motion in the y-axis. To analyse this affect the x and y components of the bubble velocity are plotted in Figure 72. The MHD forces can be seen improve the velocity of the bubble in y axis (over the bubble-induced flow field) in the initial stage of the bubble sliding process. Although, after peaking, the y-velocity gradually drops well below the non-magnetic experiment’s bubble velocity after 0.55, 0.53 and 0.35 s for $\theta = 4^\circ$, $6^\circ$ and $8^\circ$, respectively. The x-velocity’s contribution to the resultant velocity (for a combined bath flow) increases in this period of time to maintain the resultant velocity above the non-magnetic experiment. The trend of shifting velocity components is seen regardless of the inclination.

Figure 72. Bubble velocity profile (a) y-velocity (b) x-velocity
3.3.7 Conclusion

The sliding characteristics of a single bubble underneath an inclined anode surface is studied in aqueous electrolyte with and without a magnetic field. The experimental setup was modified to avoid the electrochemical generation of gas bubbles. Bubbles were created using air injection system in the electrolytic media to study the influence of magnetic field on a single bubble. Utmost care was taken to inject air at a controlled rate into the quiescent aqueous electrolyte media to avoid any bubble coalescence underneath the anode surface for each wall inclination. The behaviour of the injected single bubble was observed with a high speed camera. The bubble grows radially outward with a gradual transition to teardrop shape due to the gravity field (in absence of the external magnetic field). The bubble evolution under an external magnetic field and high current density resulted in an asymmetrically growing bubble. We observed that the bubble tends to stretch rapidly during detachment in aqueous electrolyte. In general, the sliding bubble tends to deviate from the central sliding path due to the combined influence of gravity and MHD field. The resultant flow field in the aqueous electrolyte is rather rotational than pure translational in nature. It is also interesting to see that the gas bubble holdup time or sticking to the orifice has decreased in the presence of the MHD force. This would give rise to different characteristics of anode coverage area, in terms of bubbles that nucleates in actual electrolytic cell. It is also observed that the bubble changes its shape to more complex form under an external magnetic field, which would change the associated drag force and the wake behind the bubble, and in particular the overall characteristics of the electrolytic cell.
Chapter 4

Particle Image Velocimetry Study for Electrolytic Flow

4.1 Introduction

In the previous two chapters, the bubbles sliding characteristics were studied with and without magnetic field. The bubble and electrolyte flow characteristics were found to be considerably affected by the MHD forces. The bubble sliding motion in the aqueous electrolyte shows that the size, shape, and position of the moving bubbles are unsteady and these changes may influence the Hall-Héroult process. The images that were taken in previous experiments by the high speed camera was analysed using image processing techniques. It is well-know that the bubble nucleates by displacing the surrounding fluid and changes its shape, which influence and alter the hydrodynamics of the surrounding fluid. However, it is practically impossible to know the velocity field of the surrounding fluid using only high speed imaging techniques in the previous experiments. The Particle Image Velocimetry (PIV) technique is a non-invasive measurement procedure and a powerful tool for qualitative and quantitative velocity field estimations in many fluid-flow systems. In this study the flow field around the growing, detaching and sliding bubble, underneath a downward facing anode in an aqueous electrolytic media, was investigated by PIV. The goal of the experiment was to produce a quantitative velocity field for the surrounding fluid in the aqueous cell.

4.2 Methodology

The aqueous electrolytic cell, that was modified to accommodate the air injection in previous chapter, was used for PIV measurements. The PIV system used for this experiment is a commercial DANTEC® 2D-PIV system. The PIV Laser sheet (532 nm wavelength) was generated by a Litron® 15Hz DualPower Nd:YAG Laser (1200mJ). A horizontal measuring plane was selected right beneath the anode surface as the MHD field remains dominant (parallel) across the anode surface. As the viewing area was the plane parallel to the anode surface, the laser sheet was inclined to match the anode inclination by rotating the focus module. Ideally, the laser sheet (screen) position should be cutting half-way through the bubble so that one can see the vector field around the bubble. However, PIV for a two phase flow would cause significant noise (saturation) due to the reflection from the two intersecting media. Thus, the interference of Laser sheet with the bubble was avoided by selecting a suitable measuring plane. The quantitative interest is only to compare the surrounding fluid
velocity in the presence of external magnetic field at a location near to the bubble. All measurements were taken at 10 mm below the anode, a surface parallel to anode surface, for all inclinations as shown in Figure 73. In this way, the measurement data contained the least amount of noise, while maintaining a detectable dynamic range of velocity field. A FlowSense EO 5M camera (2448 x 2050) with a Zeiss lens of 50 mm and f/1.4 was placed underneath the electrolytic cell to capture the illuminated tracer particles.

The copper anode and the gas bubble have reflective surfaces and generate some background noise due to the reflection from the laser sheet. It is to be noted that the PIV measurements have shown promising results for two phase flow in many experiments conducted in past, but the reflections from the interface of other phase (gas bubbles) are inherent in most two-phase PIV studies [87]. To mitigate this, a trial experiment was conducted with Fluorescent Polymer Particles (FPP) and a long pass filter. Rhodamine-b (Rhb) dye with a mean diameter of 26 μm was found to reduce the background noise from the anode, however, the reflection of the bubble were still visible. Additionally, ambient light was found to saturate the image and required the measurements to be conducted in a dark room. The initial velocity measurement due to the sliding of a single bubble did not induce any remarkable change in surrounding fluid hydrodynamics. Thus, the velocity of the surrounding fluid, in a location underneath the bubble was not significant, which became a major problem in terms of poor particle reaction time of the Rhodamine-b, which was used as a seeding particle for this measurement. The contamination of the bath fluid due to mixing of copper deposits at cathode and the loss of molar concentration of the electrolyte after each run requires the electrolyte to be replaced for every experiment conducted. Thus, the electrolyte was prepared with seeding particles for each
Another trial experiment was also conducted using the hollow glass spheres (HGS) of 10 μm mean diameter and a band pass filter. It was found that the HGS followed the fluid flow well and the PIV data were much better quality to capture the flow field with an acceptable low level of background noise (acceptable S/N ratio). Thus, the HGS was utilized as the tracer particles for all the experiments carried out here.

In order to obtain accurate PIV measurements, the following steps were taken in acquiring the images:

1. Size of the seeding particle to be in the range of 3-5 pixels in diameter.
3. Maximum particle displacement under 25% of the interrogation window.

The images from the camera were acquired and processed on DynamicStudio 2015a®. The raw images were masked to neglect the region of fluid outside of the anode surface. An Adaptive correlation analysis was performed to the double-frame images with an interrogation window of 64 × 64 pixels and a 50% overlap between consecutive interrogation cells. A vector masking process was applied to mask vectors for the previously user defined region (with a status code of outside, disable or rejected). Therefore, such vectors (per status code) were hidden or excluded from the analysis [88].

The UV scatter plot range validation method was used to select in a range of velocity vectors to isolate the spurious vectors. A Moving average validation method with a minimum average size of 3 × 3 was defined to replace the spurious vectors [88].

Generally, an average filter or a coherence filter was used to smooth the vector maps. The size of the averaging area, inside which individual vectors are smoothed out by the averaging vector, is defined by an appropriate value. The average filter method, filters the vector map by an arithmetic average over vector neighbours in the defined averaging area. The coherence filter was applied in some cases to the raw velocity plot to enhance the results. This method is a variation of the vector median filter that adjusts vectors that are not consistent with the dominant surrounding vectors. For a feature point (Pc) with a vector of \( \mathbf{v}_c \) (Figure 74) where all the features \( \mathbf{v}_i \) that lies inside the distance of \( S \) from the origin, Pc. The vectors \( \mathbf{v}_j \) are assumed to form a cluster due to the coherent motion [88].

\[ j \neq i \quad \text{eq. 41} \]
\[ \Delta_i = \frac{\sum_{j \neq i} \|v_i - v_j\|}{p - 1} \]  

\[ \text{eq. 42} \]

![Figure 74. Coherence filter](image)

The median vector minimizes the cumulative difference. For a cluster, the mean cumulative difference of the median velocity characterizes the spread of velocity. When the difference between \( v_c \) and \( v_{med} \) is substantial, the \( v_c \) is substituted by \( v_{med} \). In contrast, to the standard median filter which modifies the majority of the measurements, the conditional median filter only changes vectors that are likely inaccurate [88].

\[
\text{med}= \arg \min \Delta_i
\]
\[
\|v_c - v_{med}\| > \Delta_{med}
\]

\[ \text{eq. 43} \]
\[ \text{eq. 44} \]

Additionally, the individual vector maps for a defined range of time were analysed as a mean velocity vector map by the vector statistic method. This method also calculates other statistical quantities, such as, mean velocities, standard deviations, variances and covariance between different velocity components [88].

4.3 Results and Discussion

4.3.1 Bubble-induced flow field (without magnetic field)

Figure 75 shows the four phases/stages of bubble sliding: (a) just after detachment (b) halfway through the anode (c) at the edge of the anode and (d) just after release from the anode. A schematic representation of the bubble outline is shown by superposing bubble image on the vectors field. Air was injected at a controlled rate (6.2 ml/min) into the quiescent bath to create a bubble, similar to the previous experiment (chapter 3). The average bubble cross-
sectional area was 135, 69 and 45 mm\(^2\) for \(\theta = 4^\circ, 6^\circ\) and \(8^\circ\), respectively. Velocity field measurements were obtained for the surrounding electrolyte, for all inclination angles, along a plane parallel to the anode (10 mm offset from the anode surface). The Laser pulse separation time for each anode inclination ranged from 10,000 - 37,000 \(\mu s\). The velocity of the surrounding fluid is mainly caused by the bubble sliding motion as it displaces the surrounding fluid. Figure 75 shows the velocity field for \(4^\circ\) anode inclination. The vector plots show a noticeable radial velocity, ahead of the bubble nose, when bubble slides along the inclined plane by displacing the surrounding fluid. The axial component of the velocity drops off quite rapidly near the nose by pushing the surrounding fluid radially outward as mentioned above. It is also clear from the vector plots that the surrounding fluid begins to flow in a reverse direction over the bubble surface (body) opposing the bubble motion. The velocity over the bubble increases rapidly during detachment due to change in the bubble stretching ratio. It is also interesting to see that the surrounding fluid is pulled inward towards the rear side (tail) of the bubble due to the formation of wake behind the bubble.

Figure 75. Bubble-induced flow at four stages for \(\theta = 4^\circ\)
Figure 76 to Figure 78 show the contour plots of velocity magnitude and the streamlines at two selected bubble locations ($\Delta t = 0.125$ s) for all inclinations of anode. The figures clearly indicate that the velocity of surrounding fluid changes the direction at frontal and rear sides of the sliding bubble. On each flow field plot there are two distinct locations at the tail and nose end of the bubble where the velocity is measured to be nearly zero. The flow at these locations are primarily along the $z$-axis, thus, invisible to the 2D PIV measurements. The average velocity of the entire electrolyte (bath) was calculated for the four different phases/stages and plotted for all inclination angles in Figure 79(a). Due to the reduction in bubble height with rise in inclination, the distance between the measured plane and the top of the bubble grows with higher inclination angles. This is especially noticeable for the $\theta = 8^\circ$ where the trend of higher inclination to higher bath velocity is not applicable as shown in Figure 79(b).

![Figure 76. Bath velocity field for $\theta = 4^\circ$ at 0.125 s intervals after detachment](image)

![Figure 77. Bath velocity field for $\theta = 6^\circ$ at 0.125 s intervals after detachment](image)
Figure 78. Bath velocity field for $\theta = 8^\circ$ at 0.125 s intervals after detachment

Figure 79. Bubble-induced flow field (a) at each phase of bubble sliding (b) mean bath velocity
4.3.2 MHD-induced flow field

Experiments were carried out to measure velocity by using a steady 3 ampere current ($\approx 376 \text{ A/m}^2$) into electrolyte with the static magnetic field at one end of the cell. Initially, the velocity measurement were carried out without injecting any bubbles to quantify the MHD induced flow field of the electrolyte. PIV measurements were obtained for a Time Between Pulse (TBP) of 4,500 $\mu$s at a maximum trigger rate of 8 Hz. It is expected that the Lorentz field due to the current in the electrolyte and magnetic field would induce a flow in the electrolyte (bath) with maximum velocity near the sidewall of the cell. Figure 80 shows the velocity contour plots, streamlines and velocity vectors at $4^\circ$ anode inclination. It was found that the velocity at the right side of the anode, i.e. in the vicinity of external magnet, was maximum, which is obvious because of the existence of higher Lorentz field. It was surprising to see that the velocity at some locations were as high as 0.07 m/s. However a uniform velocity was not observed due to the imperfection on the anode surface that changes the local current density and thereby the variation in Lorentz field.

The bath velocities (underneath the anode) are determined at three regions (as shown in Figure 81) of the measuring plane to provide an average velocity for each region. Figure 82 shows that the velocity at the edge is higher for all inclinations while the middle region of the anode has the lowest velocity. It is interesting to see a counter-clock-wise rotational flow throughout the entire plane (as shown in Figure 80), which may be described as Lorenz force driven forced convention or MHD induced flow, is established in the cell prior to the bubble nucleation. Mutschke, et al. [89] have reported similar counter-clockwise flow resulting from Lorentz driven convective forces. It is important to note that the order of magnitude of MHD force in the electrolyte is significantly higher than the bubble-induced flow field that was measured previously. The increase in anode inclination resulted in a higher mean velocity in the electrolyte (shown in Figure 83), which may be attributed to the shifting of current density towards the leading edge of the anode.
Figure 80. MHD flow field at $\theta = 4^\circ$

Figure 81. Three zones of the anode
Figure 82. MHD-induced bath velocity at (a) $\theta = 4^\circ$ (b) $\theta = 6^\circ$ (c) $\theta = 8^\circ$

Figure 83. Mean bath velocity for MHD flow field as a function of anode tilt angle.
Experiments were also carried out to quantify the velocity of the electrolyte flow in presence of the magnetic field with electrical current and continuous injection of bubbles (similar to Chapter 3). Measurements were conducted at a time separation of 4,500 µs and maximum trigger rate of 8 Hz. Figure 84 shows that a dominant MHD flow overshadows the flow field pattern induced by the sliding bubble. The flow, near the edge of the anode, is seen to be more stable, showing a boundary layer growth at the side wall of the cell. Figure 85 shows the average velocity for the three zones, which depicts a similar pattern/trend as shown in Figure 82. The average bath velocity for all four phase/stages (Figure 86) show that the bubble injection and MHD field together give rise to a higher velocity for all inclinations than simply MHD driven flow. Thus, the bubble induced flow has an impact on the overall bath flow characteristics.

Figure 84. Combined flow field at $\theta = 4^\circ$
Figure 85. Combined flow bath velocity at (a) $\theta = 4^\circ$ (b) $\theta = 6^\circ$ (c) $\theta = 8^\circ$

Figure 86. Mean bath velocity as a function of inclination angle
4.4 Dimensionless Analysis

The first step towards a more quantitative approach is a dimensionless analysis because it allows to reduce the number of variables needed for physical problem. Although the PIV measurements provide a quantitative velocity field for the aqueous electrolytic cell, it is necessary to relate the velocity with other associated parameters to provide a general characteristics of Hall-Héroult cell.

Alam, et al. [6] utilized five dimensionless numbers as proposed by Zhang, et al. [90] to maintain the dynamic similarity for a 1:4 scale aluminium reduction cell at isothermal condition. They used modified Froude, Weber, Reynolds, Eötvös and Morton numbers. The Froude number gives a comparison of inertial forces to the gravitational forces, Weber number gives the ratio of inertial forces to surface tension forces [91, 92]. While the Reynolds number is a ratio of inertia forces to viscous forces [93].

\[
\text{Modified Froude Number (Fr)} = \frac{\rho_g q^2}{(\rho_1 - \rho_g)gL} \quad \text{eq. 45}
\]

\[
\text{Modified Weber Number (We)} = \frac{\rho_g q^2 L}{\sigma} \quad \text{eq. 46}
\]

\[
\text{Modified Reynolds Number (Re)} = \frac{\sqrt{\rho_1 - \rho_g}qL}{\mu} \quad \text{eq. 47}
\]

Where, \( \rho_1 \) and \( \rho_g \) are liquid and gas density, respectively, \( L \) is the characteristic dimension, \( q \) is the gas generation rate for a unit surface area, \( \sigma \) and \( \mu \) are the surface tension and the dynamic viscosity of the surrounding fluid, respectively [6, 38].

Similarly the Morton number (\( M_o \)) and Bond number (\( B_o \)) relates gravitational forces to surface tensional forces as given below [4, 94].

\[
M_o = \frac{g\mu^4(\rho_1 - \rho_g)}{\rho_1^2 \sigma^3} \quad \text{eq. 48}
\]

\[
B_o = \frac{g(\rho_1 - \rho_g)L^2}{\sigma} \quad \text{eq. 49}
\]

These dimensionless parameters are used to characterise the bath flow phenomena in the electrolytic cell. For example, the size and shape of a moving bubble in a viscous media can be characterized by the Bond number and Morton number [6]. While the Eötvös number is
used to describe the bubble diameter before detachment. Thus, it is important to know the
graph. geometry and electrolyte properties for any scaling-analysis. Table IV shows the comparison
of the variables of an industrial cell, physical model used by Alam, et al. [6] and the
experimental rig used in the present work. The bubbles formed in the revised physical model
is similar to an industrial cell, in terms of, Eotvos number and Morton number.

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Electrolyte</td>
<td>Cryolite</td>
<td>0.28M CuSO₄ + 20% H₂SO₄</td>
<td>0.32M CuSO₄</td>
</tr>
<tr>
<td>Electrolyte density (kg/m³)</td>
<td>2100</td>
<td>1195[95]</td>
<td>1051.1[96]</td>
</tr>
<tr>
<td>Electrolyte surface tension (mN/m)</td>
<td>129</td>
<td>98.7[97]</td>
<td>73.4 [98]</td>
</tr>
<tr>
<td>Electrolyte viscosity (kg/ms)</td>
<td>0.00251</td>
<td>0.0011[95]</td>
<td>0.00122[96]</td>
</tr>
<tr>
<td>Bubble diameter before release (mm)</td>
<td>11 to 13</td>
<td>2 to 31</td>
<td>13.72, 9.57 &amp; 7.92 (θ = 4°, 6° &amp; 8° respectively)</td>
</tr>
<tr>
<td>Anode length (m)</td>
<td>1.35[37]</td>
<td>0.35</td>
<td>0.095</td>
</tr>
<tr>
<td>Modified Froude</td>
<td>1.16 x 10⁻¹⁰</td>
<td>1.43x10⁻¹¹</td>
<td>2.52x10⁻¹³</td>
</tr>
<tr>
<td>Eötvös number</td>
<td>19.3 to 27</td>
<td>0.5 to 114</td>
<td>26.4, 12.8 &amp; 8.8 (θ = 4°, 6° &amp; 8° respectively)</td>
</tr>
<tr>
<td>Morton number</td>
<td>8.64x10⁻¹¹</td>
<td>1.25x10⁻¹¹</td>
<td>5.24x10⁻¹¹</td>
</tr>
</tbody>
</table>

The anodic gas bubbles are highly resistive electrically, thus, do not experience the effect of
MHD forces directly. However, there is a strong interplay between the pressure, viscosity,
density, surface tension, gravity and the conductivity of the surrounding fluid with MHD field
[99]. In this section, an attempt is made to understand the effects of MHD forces on the
electrolytic flow by deriving the dimensionless numbers taking all the parameters into
consideration. It was also proposed to relate the dimensionless parameters, particularly the
MHD inaction parameter with PIV velocity measurements. In a two-phase flow system under
isothermal conditions the velocity of a sliding bubble in an electrolytic cell can be expressed
as shown in eq. 50.

\[ v_b = f(\rho, \rho_b, \mu, L, d_b, \gamma, g, B, \sigma, Q, V) \]

\[ \text{eq. 50} \]
Where $\rho$ and $\rho_b$ are the density of the electrolyte and the gaseous phase, respectively; $\mu$ is the dynamic viscosity of the electrolyte; $L$ is the characteristic length of the anode; $d_b$ is the bubble diameter; $\gamma$ is the surface tension of the electrolyte; $g$ is the gravity; $B$ is the magnetic field; $\sigma$ is the conductivity; $Q$ and $V$ are the gas formation rate and bulk velocity of the electrolyte, respectively.

The functional, $f$, shown in eq. 50, contains 12 dimensionally homogeneous variables and 4 reference variables (Mass, Length, Time and Ampere) [100]. The Buckingham Pi ($\pi$) Theorem is used to decide the number of independent dimensionless parameters needed to fully describe the behaviour of this system, where $n - j = 12 - 4 = 8$. A summary of the variables with the dimensions can be given as follows:

<table>
<thead>
<tr>
<th>Factors affecting bubble velocity</th>
<th>Units</th>
<th>MLTA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density of the liquid ($\rho$)</td>
<td>kg/m$^3$</td>
<td>ML$^{-3}$</td>
</tr>
<tr>
<td>Density of the gas ($\rho_b$)</td>
<td>kg/m$^3$</td>
<td>ML$^{-3}$</td>
</tr>
<tr>
<td>Dynamic viscosity of the liquid ($\mu$)</td>
<td>kg/sm</td>
<td>MT$^{-1}$L$^{-1}$</td>
</tr>
<tr>
<td>Characteristic length of the anode ($L$)</td>
<td>m</td>
<td>L</td>
</tr>
<tr>
<td>Bubble diameter ($d_b$)</td>
<td>m</td>
<td>L</td>
</tr>
<tr>
<td>Surface tension of the liquid ($\gamma$)</td>
<td>N/m</td>
<td>MT$^{-2}$</td>
</tr>
<tr>
<td>Acceleration due to gravity ($g$)</td>
<td>m/s$^2$</td>
<td>LT$^{-2}$</td>
</tr>
<tr>
<td>Magnetic field ($B$)</td>
<td>N/Am</td>
<td>MT$^2$A$^{-1}$</td>
</tr>
<tr>
<td>Conductivity ($\sigma$)</td>
<td>A$^2$s/kg m$^3$</td>
<td>A$^2$T$^3$L$^{-3}$M$^{-1}$</td>
</tr>
<tr>
<td>Gas formation rate ($Q$)</td>
<td>m$^3$/s</td>
<td>L$^3$T$^{-1}$</td>
</tr>
<tr>
<td>Bulk velocity of the electrolyte ($V$)</td>
<td>m/s</td>
<td>LT$^{-1}$</td>
</tr>
</tbody>
</table>
The number of repeating variables equal the number of reference variables. For this analysis, the density of the gas phase, dynamic viscosity of the electrolyte, magnetic field and bubble diameter terms are chosen as the repeating variables to form the \( \pi \) terms by combining with the other remaining variables. For instance, the dimensional product with surface tension can be expressed as shown in eq. 51.

\[
\pi = \rho_b \mu^d B_c^e, d_b^d, \gamma
\]

\text{eq. 51}

The four exponents \( a, b, c \) and \( d \) are determined by equating the power of dimensional quantities and the dimensionless parameter is shown in eq. 52.

\[
\pi = \frac{\rho_b d_b \gamma}{\mu^2}
\]

\text{eq. 52}

A similar procedure gives rise to set of dimensionless parameter, which can be expressed as:

\[
\frac{\rho_b v_b d_b}{\mu} = f \left( \frac{\rho}{\rho_b}, \frac{L}{d_b}, \frac{\rho_b d_b \gamma}{\mu^2}, \frac{\rho_b^2 d_b g}{\mu^2}, \frac{\sigma B^2 d_b^2}{\mu}, \frac{\rho_b Q}{\mu d_b}, \frac{\rho_b V d_b}{\mu} \right)
\]

\text{eq. 53}

Where,

\[
\pi_1 = \frac{\rho_b v_b d_b}{\mu}; \quad \pi_2 = \frac{\rho}{\rho_b} = \rho^*; \quad \pi_3 = \frac{L}{d_b} = L^*; \quad \pi_4 = \frac{\rho_b d_b \gamma}{\mu^2};
\]

\[
\pi_5 = \frac{\rho_b^2 d_b g}{\mu^2}; \quad \pi_6 = \frac{\sigma B^2 d_b^2}{\mu} = Ha^2; \quad \pi_7 = \frac{\rho_b Q}{\mu d_b}; \quad \pi_8 = \frac{\rho_b V d_b}{\mu}
\]

Most common dimensionless number such as Reynolds number can be seen as:

\[
Re_b = \frac{\rho_b v_b d_b}{\mu}
\]

\text{eq. 54}

\[
Re_M = \frac{\rho_b V d_b}{\mu}
\]

\text{eq. 55}

Where \( Re_b \) and \( Re_m \) are the Reynolds number based on characteristics of the bubble and MHD induced flow respectively. Figure 87 and Figure 88 plots \( Re_b \) and \( Re_m \) vs. time to show the characteristics of the analogous model used in this experiment.
Other conventional dimensionless parameters can be derived by combining the above \( \pi \) groups in many different ways. For example, Bond number, which is a ratio of gravitational to surface tension force, can be expressed as a combination of \( \pi_5 \) and \( \pi_4 \) as shown below:

\[
Bo = \frac{\pi_5}{\pi_4} = \frac{\rho g d_b^2}{\gamma}
\]

The most important non-dimensional parameters describing the influence of magnetic field are the Hartmann number and the magnetic interaction parameter (Stuart number) [99], which can be written as:

\[
Ha = Bd_b \sqrt{\frac{\sigma}{\mu}} = \frac{\text{Lorentz Force}}{\text{Viscous Shear Force}}
\]

\[
N = \frac{\sigma B^2 L}{\rho V} = \frac{Ha^2}{Re} = \frac{\text{Lorentz Force}}{\text{Inertia Force}}
\]

Figure 87. Modified Reynolds number (bubble) as a function of time
The Hartmann number (eq. 57) is the ratio of electromagnetic forces to viscous forces where it measures how strongly the fluid flow changes due to the magnetic field [101]. The Stuart number (eq. 58) defines the ratio of magnetic forces to the inertial forces [102]. Figure 89 shows the dominance of Lorentz force growing over the viscous shear and inertia force as the bubble moves towards region of higher magnetic field and current density. Although, the inertia force is stronger than the viscous shear force.

Shin and Kang [103] reported on the effect of uniform magnetic field on the shape of rising incompressible gas bubble in a liquid metal. The numerical study predicted that the rising bubble would experience an elongation in the direction of the magnetic field. Where elongation increased monotonously with the rise of Stuart number. Shibasaki, et al. [85] also reported on similar bubble deformation to a long spheroidal shape. Additionally, they predicted a deceleration of the fluid flow (MHD braking effect [104]) for Ha>75. The gradual growth of MHD forces characterised by the Hartmann number and Stuart number correlates with the changes seen in the bubble morphology in Chapter 3. Although the analysis given in this section, due to the low electrical current and magnetic field strength, is limited by the scale of the physical model but is appropriate characterising any scale-up study with given dynamic similarity.
A PIV measurement was carried out for quantify the velocity of electrolyte for all inclinations. Velocity data were taken along 2D plane parallel to the anode surface. The main objective of this chapter was to validate, examine and quantify the two sources of bath momentum (bubble and MHD) with respect to the anode inclinations. The bubble-induced flow field was found to be dependent on the bubble sliding motion. However, the effect of the inclination on the average bath velocity was found not be consistent on the plane where velocities were
measured. This is mainly attributed to the shape change of the bubble that occurs during the sliding motion. The MHD-induced-flow field was found to drive the entire region of fluid domain. It is interesting to see a counter-clock-wise rotational flow throughout the entire plane which may be described as Lorenz force driven forced convention. Moreover, the magnitude of the mean bath velocity was significantly higher than the bubble-induced flow. The effect of higher anode inclination was found to improve mean bath velocity at higher inclinations, which is interesting, since the only significant change that is expected is the shift in current density towards the leading edge of the anode.

A dimensionless analysis was also carried out considering all variables that are relevant to Hall cell. The velocity measured from PIV was used to quantify the dimensionless parameters. The most important non-dimensional parameters describing the influence of magnetic field are the Hartmann number and the magnetic interaction parameter which were derived. The characteristics of the MHD induce flow were plotted for all inclinations to show the influence of magnetic field. The Hartmann number and Magnetic interaction parameter show that as the bubble moves towards the anode edge, the dominance of the MHD forces grows over the viscous shear force.
Chapter 5

General Discussion

The characteristics of gas bubbles forming underneath a surface play a vital role in many electrolysis processes. In particular, the aluminium reduction process where bubbles nucleate under strong magnetic field and current density. The motivation behind this study was to quantify and demonstrate importance of MHD forces to the electrolytic cell process by studying the behaviour of bubbles and flow field measurements. The aspects of bubble formation, sliding and coalescence affects the bubble-induced resistance within the cell. More precisely, the anodic gas bubble size, morphology and holdup time contributes to the complex electrical resistance that bubbles induce within the cell. Furthermore, the motion of the bubbles is well known to add momentum to the bath, which is necessary to the homogenisation of temperature and bath concentration and the removal of other bubbles. However, the high temperature and highly corrosive environment of the Hall-Héroult cell severely limits the study of the bubble behaviour in this environment. Thus, many researchers have developed laboratory physical models that replicate the environment of the cell with simplifications that enable the study of the bubble behaviour under various parameters. The previous studies have been able to achieve geometric similarity with some dynamic similarity. Although they have only been able to observe and measure the bubble characteristics with only a bubble-induced flow field. It is well known that the high electrical current in the conductive fluids together with intense magnetic field that is generated by the external cathode busbars interact to form Lorentz forces. Despite the strong impact of electromagnetic forces on the pressure, viscosity, density, surface tension and gravity field in the electrolyte bath, which indirectly affect the anodic gas bubbles, no research has been conducted into the effect of combined flow field and quantitatively establish which source of bath momentum is dominant.

The bubble-induced flow field and MHD-induced flow field have two widely different mechanisms that engender bath flow. The bubble-induced flow field has been studied under various geometric parameters and boundary condition with experimentation and numerical analysis over the past decades. Under the branch of bubble-induced flow, two distinct type of bubble formation methods have been investigated. The earliest and most extensive has been the air-injection method and more recently the electrolytic cell model. However, both traditional physical modelling techniques are only able to study under bubble-induced flow. While in an actual cell, the flow has a combined effect, which has been studied only in numerical models that have been predicted to be significantly different to that of the bubble-induced flow, in terms of the magnitude and flow pattern. This fact emphasises the importance
of combined flow field and contests the idea of neglecting the MHD forces as insignificant component in the bath flow. It changes the observations and conclusions of bubble characteristics that have been made in the past without MHD forces.

The present work, therefore, addresses the bubble behaviour under strong magnetic fields where the electrolytic bath flow is a combined flow field. The main difficulty lies with attaining a similar current distribution to an industrial cell in a scaled-down model using an aqueous electrolytic solution. Therefore, the aim was to achieve an inclined current path in the electrolyte while trading-off geometric similarity. The intension of the Chapter 2 was to develop a laboratory scaled physical model that incorporates both sources of bath momentum and to study its impact on the electrolytically forming bubbles. This is the first of study of its kind and is not exhaustive. The high-speed videography showed a distinctive and widely different bubble phenomenon exclusive to the observations made in this study. The formation of a combined flow field and the impact on sliding bubbles at various wall inclinations are novel aspects which are addressed in the present work. Apart from the knowledge that is applicable to the Hall-Héroult process, the fundamental changes in the bubble behaviour can contribute to other electrolytic processes where bubble formation is naturally occurring. The emphasis was on the characteristics of bubbles, such as, spread of nucleation sites, bubble size, bubble morphology, sliding velocity and the sliding path with different anode inclination. The comparison of these fundamental aspects of bubbles under the electromagnetic forces will allow us to better optimize the electrolytic process and understand the impact of accessories that aim to reduce the effect of Lorentz forces.

In Chapter 2, the linearly moving bubbles were observed to move parallel to the anode edge under an external magnetic field. Three distinct bubble flow patterns were observed, where the bubble behaviour was based on the proximity to the magnetic field and current density. At the predicted high MHD flow zone, the bubbles were moving with considerably high velocity, parallel to the anode edge. The strong electrolyte flow in this zone sweep all bubbles, thus impeding any significant formation of bubbles in this region. The majority of the bubbles that are moving in this region originated at zone 2. The bubble formed in zone 2 nucleate with a shorted bubble to bubble distance with a sluggish swarm flow that resembles a rotational bubble path. Some isolated bubbles were found to nucleate at zone 3. Nevertheless, the introduction of the MHD forces were found to considerably improve the sliding velocity on all regions of the anode. A main drawback of this study was the small size of the nucleated bubbles which made the observation of shape change difficult, especially with higher inclination. Also, the bulk bubble behaviour made it difficult to locate any single bubble
undergoing through all different stages of formation, detachment and sliding underneath the anode surface. Joule heating was kept to a minimum by the low amount of electrical current that was supplied to the cell.

To study the sliding bubbles in a fundamental level, the effect of bubble swarm flow was avoided. This was accomplished by altering the electrochemical process to stop the generation of gas bubbles, while air bubbles were injected at a single submerged orifice. It was found that the effect of MHD forces grew as the bubble moved towards the edge of the anode. Similarly, to the electrolytic bubbles, the drifting velocity of the injected bubbles were enhanced for all inclination angles. Additionally, the bubble experienced complex changes in its shape as it entered regions of strong MHD forces and this effect was more apparent with higher inclination. The flow field measurements showed that this was in part due to the improvement in bath velocity with higher inclination. Although, the effect of inclination on the bubble induced flow field velocity did not show a consistent trend. This was primarily due to the fact that the measuring plane is fixed at 10 mm below the anode, while the height of the bubble reduced with higher inclination. Thus, the effect of the bubble at the examined plane grows lesser. Nevertheless, the mean bath flow velocity was higher for the combined flow, in comparison, to the MHD induced flow. Therefore, both components of the bath flow is important in determining the resulting flow.

The bubbles formed in the revised physical model (Chapter 3) were found to be similar to an industrial cell, in terms of, Eötvös number and Morton number. The derived MHD interaction parameter characterised a step change of the anode tilt angle. The measured flow fields quantified and provided direct evidence that MHD convection can improve the overall bath velocity, which was shown to improve the detachment and sliding velocity of the gas bubbles and it also may favour an optimized chemical reaction.

The results obtained in this study shows dramatic change in the fundamental behaviour of bubbles formed underneath a surface. The major limitation in the experiment model was the aqueous electrolyte, which has a very low electrical conductivity. For a fixed size of electrode, a fixed amount of current injection is possible. Thus, it is practically impossible to varying the current and subsequently the magnetic field. The work done here can add to the interest of the scientific community and to further advance the development of the aluminium reduction cells. However, the direct transfer of the conclusions to an industrial Hall-Héroult cell is not possible due to the complexity of the actual cell.
5.1 Conclusion

An experimental study to investigate the impact of combined flow field on the behaviour of bubbles under a plane was conducted successfully with an emphasis on inclination angle, bubble generation method and bath flow field. The key findings from the present work are:

a. The presence of MHD forces changed the bubble sliding behaviour fundamentally. The linear bubble sliding path in a bubble-induced flow field (orthogonal to the anode edge) change to a rotational path towards the edge. This phenomenon is a distinguishable and unique. Anode tilt angle was found to influence the bubble sliding path under magnetic field.

b. The combination of bubble-induced and MHD induced flow field was able to improve the bubble creeping velocity by a considerable margin on both injected and electrolytically generated bubbles.

c. Bubbles injected in the high MHD zone grew asymmetrically with a shorter holdup time and experienced complex uneven shape change.

d. The bath velocity of the EMF induced flow field is significantly higher than the bubble-induced flow field. Also, the higher anode inclination improves the mean bath velocity of the EMF flow field.

e. The average bath velocity of the combined flow field was higher than the EMF only flow field, however, due to selected measuring plane the effect of inclination was not consistent.

5.2 Recommendations for future work

a. Under a magnetic field, conducting a study to examine the effect of different gases (lower density) at a higher gas generation rate in a porous anode to match the Modified Froude number.

b. Examining the difference in bath flow field between the injected bubble and electrolytic bubble in a combined flow field.
Reference List


