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Abstract

It is known from aeromagnetic surveys from Venezuela, China and the U.S.A. that it is common to find geomagnetic anomalies over oil fields. It has been suggested that hydrocarbon reservoirs, through upward seepage, create physical and geochemical conditions in soils and rocks sitting above them that are conducive to the formation of secondary ferrimagnetic minerals such as magnetite and greigite. In near-surface samples drilled in Venezuela it has been observed that these minerals form aggregates of roughly spherical grains called *framboids*. While the specific physical and geochemical processes involved in the genesis of these minerals is an active research topic, I will focus in studying the domain structure and hysteresis parameters of these minerals via numerical simulation of the *micromagnetic* equations. Solving the micromagnetic problem can be done either by minimising the total Gibbs free energy which is a sum of energies associated with different phenomena in a ferromagnetic material or by solving the dynamical equation for the magnetic moments, that is, the Landau-Lifshitz-Gilbert equation (LLGE). I will use a finite element method based on the general-purpose package collection for automated solutions of partial differential equations *FEniCS* to solve the LLGE as well as an energy minimising routine, specifically a conjugate gradient method. My aim is to find the magnetic response of these mineral aggregates measured in a given sample by standard rock-magnetic and palaeomagnetic techniques. As a first approach to the problem, before scaling up the simulations, I have conducted simulations of hysteresis loops and zero-field domain structure of octahedral-shaped single grains of greigite and compared these with octahedral grains whose corners have been chopped. These shapes are typical morphologies of greigite and so, these simulations constitute an improvement over previous finite difference models that could only study somewhat unrealistic shapes like cubes and rectangular prisms.

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Chapter 1

Introduction

1.1 Motivation and Objectives

It has been established via airborne magnetic surveys in the U.S.A. (Donovan et al., 1979) that magnetic contrasts—that is, “magnetisation that is different from background magnetisation and which may give rise to mappable magnetic anomalies detectable by conventional magnetometry” (Machel and Burton, 1991)—are a common feature of hydrocarbon reservoir sites. Donovan et al. (1979) suggested that these magnetic anomalies are caused by near-surface magnetic minerals (specifically, magnetite) induced by upward hydrocarbon seepage from the underlying hydrocarbon reservoir. Further studies by Donovan et al. (1984), Reynolds et al. (1993) and Elmore et al. (1993) in the U.S.A., Díaz et al. (2000), Costanzo-Alvarez et al. (2006), Guzmán et al. (2011), González et al. (2002) in Venezuela and Liu and Liu (1999), Liu et al. (2004) and Liu et al. (2006) in China have provided strong evidence for a genetic relationship between the magnetic contrasts produced by ferrimagnetic minerals near the surface and the underlying reservoir. These investigations confirm the original hypothesis of Donovan et al. (1979) that the reducing environment caused by the upward seepage from the reservoirs is conducive to the formation of magnetic minerals—such as magnetite and other Fe-oxides, and greigite and other Fe-sulfides—and/or the destruction of minerals such as hematite (Machel and Burton, 1991) and thus further the case for using a combination of aeromagnetic surveying and

rock-magnetic measurements of soils and rocks for cheap hydrocarbon prospecting as proposed by Donovan et al. (1984).

Though the discussion on the exact mechanism for the formation of these minerals is ongoing, Machel and Burton (1991) have identified two primary agents for the precipitation of magnetic minerals under the influence of hydrocarbon seepage. At higher temperatures and thus higher depths they propose chemical processes as the main factor while at shallower depths and lower temperatures it is argued that microbial sulfate-reducing processes are playing the larger role. Machel and Burton (1991) also emphasised the difficulty in linking a magnetic anomaly to a process of hydrocarbon seepage because the precipitation of magnetic minerals can cause positive or negative anomalies—that is, peaks or dips in the geomagnetic field and/or the magnetic susceptibility of the soils. Nevertheless, careful analysis of the local conditions can result in the successful application of rock-magnetic measurements to hydrocarbon exploration (see Liu et al. (2006) and Donovan et al. (1984)). Magnetization of oil bearing rocks can also be used to assess the quality of oil as discussed by Emmerton et al. (2013).

It was recognized by Reynolds et al. (1993) that in some cases iron sulfides may be more important to the magnetic contrasts and thus to the identification of prospective oil-producing fields than iron oxides. Particularly, greigite has been identified as an authigenic mineral of the utmost importance in the Simpson oil field in Alaska (Reynolds et al., 1993). Greigite is an iron sulfide (Fe_3S_4) that can be thought of as the sulfur equivalent of the iron oxide magnetite (Fe_3O_4) as they have the same crystal structure only with sulfur replacing oxygen. Like magnetite, it is highly magnetic. Nevertheless, since it is thought to be unstable, its importance as a palaeomagnetic recorder has not been as readily realized as that of magnetite. Also, its magnetic parameters were poorly understood until the work of Chang et al. (2008) who by synthesising highly pure greigite were able to measure the critical magnetic parameters. Muxworthy et al. (2013) made use of the new accurate measurements of greigite to simulate greigite grains and the effect of intergrain interactions.

Liu et al. (2006) has proposed that magnetic mineral grains that are linked to hydrocarbon seepage have sizes $\sim 25\text{nm}$ and thus generally in the single domain (SD) range. In terms of

morphology, it has been repeatedly found (see Aldana et al. (1999) and references therein) that roughly spherical grains of magnetite and/or greigite assemble in raspberry-shaped aggregates called framboids (from the french *framboise* meaning raspberry). It is my interest to model these structures via micromagnetics to investigate their domain structure and behaviour in routine rock-magnetic and palaeomagnetic measurements that could also correlate with the size of the aggregates. It is an open question how the intergrain interactions will affect the otherwise simpler problem of modelling single grains, however Muxworthy et al. (2013) have made a strong case for the importance of intergrain magnetostatic interactions. Further understanding of framboidal aggregates of greigite or magnetite could aid in developing future oil exploration techniques that are cheaper than more conventional techniques vastly used today.

Chapter 2

Background Theory

2.1 Magnetism and matter

2.1.1 Fundamentals of magnetism

The magnetic field, just like the electric field, is defined by its effect on a *test charge*, namely the Lorentz force:

$$\vec{F} = q\vec{E} + q\vec{v} \times \vec{B}, \quad (2.1)$$

where q is the electrical charge, \vec{v} its velocity and \vec{E} and \vec{B} the electrical and magnetic field (although for historical reasons \vec{B} is called the *induction* field). Clearly the force a magnetic field exerts on a moving electrical charge is perpendicular to both its velocity and to the field itself. From the Lorentz law it is readily seen that the unit of \vec{B} is $\frac{\text{N}}{\text{C} \cdot \frac{\text{m}}{\text{s}}}$, this physically meaningful unit is called Tesla, with symbol T.

The description of magnetic fields is analogue to that of electrical fields. Although, unlike the situation in electricity, there are no magnetic charges, only magnetic dipoles. A more thorough analysis reveals the origin of magnetism is electrical: an electrical current (either a flow of electrons or a single electron) produces an induction field \vec{B} . The induction due to a wire carrying a current I (generally varying along the path) at point \vec{r} can be calculated from the

Biot-Savart law:

$$\vec{B}(\vec{r}) = \frac{\mu_0}{4\pi} \int_C \frac{I d\vec{l} \times \vec{r}}{r^3}, \quad (2.2)$$

where $d\vec{l}$ is a differential element of length along the wire in the direction of the current. The integral is carried out along a line, usually but not necessarily a closed curve. Thus, on a fundamental level it is correct to understand magnetism as an electrical phenomenon.

When describing magnetic fields the *magnetic induction* \vec{B} and the *magnetic field* \vec{H} are used. These denominations are of historical character and it has been proven that the true fundamental field is the induction field \vec{B} (for a thorough discussion of why this is so see Feynman et al. (1964)) while \vec{H} represents contributions to the field by macroscopic currents. Other contributions to the magnetic field are due to atomic dipoles and circular currents in a medium. In a vacuum, \vec{B} and \vec{H} coincide in direction as there are no atomic magnetic dipoles present. In the SI the magnetic and induction field differ by a scalar factor μ_0 , the *magnetic constant*, also known as the vacuum permeability or permeability of free space:

$$\mu_0 = \frac{B_{vacuum}}{H} = 4\pi \cdot 10^{-7} \frac{\text{V} \cdot \text{s}}{\text{A} \cdot \text{m}}. \quad (2.3)$$

Although the names vacuum permeability and permeability of free space are still widespread it is preferable to use the name magnetic constant since it reflects the fact that it's a defined value and not a measurement.

In general, we are not interested in describing magnetic phenomena in a vacuum, rather it is more interesting and important to consider the presence of matter. A material is composed of atoms, which individually may hold a permanent magnetic dipole moment $\vec{\mu}$ (with units $\text{A} \cdot \text{m}^2$). The *magnetisation* vector field \vec{M} is the spatial average of a myriad of these individual atomic dipole moments over a suitable volume. Therefore, \vec{M} accounts for the contribution of atomic magnetic moments to the total field. In the SI units we have

$$\vec{B} = \mu_0(\vec{H} + \vec{M}); \quad (2.4)$$

immediately we see that both \vec{H} and \vec{M} have the same units, the Ampere per meter $\frac{\text{A}}{\text{m}}$.

Mineral	Magnetic susceptibility (per unit mass)
	($10^{-8}\text{m}^3/\text{kg}$)
<i>Diamagnetic</i>	
Quartz (SiO_2)	-0.62
Orthoclase feldspar (KAlSi_3O_8)	-0.58
Calcite (CaCO_3)	-0.48
Forsterite (Mg_2SiO_4)	-0.39
Water (H_2O)	-0.90
<i>Paramagnetic</i>	
Pyrite (FeS_2)	30
Siderite (FeCO_3)	123
Ilmenite (FeTiO_3)	100–113
Orthopyroxenes ($(\text{Fe,Mg})\text{SiO}_3$)	43–92
Fayalite (Fe_2SiO_4)	126
Intermediate olivine ($(\text{Fe,Mg})_2\text{SiO}_4$)	36
Serpentine ($\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_4$)	≥ 120
Amphiboles	16–94
Biotites	67–98
Illite (clay)	15
Montmorillonite (clay)	14

Table 2.1: Magnetic susceptibilities of diamagnetic and paramagnetic materials. Reproduced from Dunlop and Özdemir (1997).

2.1.2 Diamagnetism and paramagnetism

Magnetism in matter can be broadly categorised into three phenomena: diamagnetism, paramagnetism and ferromagnetism. Diamagnetism and paramagnetism are of little importance to us so we will only briefly describe them.

Diamagnetism is a property of all matter. It is the smallest effect and is a tendency of a material to oppose an external magnetic field. An external magnetic field exerts a Lorentz force on the bound electrons that causes them to precess like a gyroscope. This is called Larmor precession and is equivalent to an electric current producing a magnetic moment in the direction opposed to the external \vec{B} field. Water is a common example of a highly diamagnetic material.

Paramagnetism is a partial alignment of the permanent atomic magnetic moments of the atoms in a material with an external \vec{B} field. It is only thermal noise that prevents a perfect alignment of the atomic magnetic moments with the external field, therefore this is a highly temperature dependent phenomenon. Nevertheless, at ordinary temperatures, paramagnetism outweighs

diamagnetism by a factor greater than 10.

In both diamagnetism and paramagnetism a magnetisation is induced by an external field. The rate at which the magnetisation is acquired with respect to the applied field is called the *magnetic susceptibility* $\chi = \frac{d\vec{M}}{d\vec{H}}$, in general a tensor. Table 2.1.2 (reproduced from Dunlop and Özdemir (1997)) lists susceptibility values for some diamagnetic and paramagnetic materials.

2.1.3 Ferromagnetism

A common theme in diamagnetism and paramagnetism is that an external field induces a temporary magnetisation in the material; once the external field is removed the material loses its magnetisation. Also, at ordinary temperatures, only extremely high external fields of the order of 100 Tesla (for a sense of scale, the Earth's magnetic field is of the order of 10^{-4}T) can induce in a paramagnetic material a saturation magnetisation—that is, a magnetisation that results from all the atomic magnetic moments aligning in exactly the same direction. A ferromagnetic material like Iron or Nickel is fundamentally different in both these aspects. It only takes external fields of the order of 0.1 Tesla to achieve a saturation magnetisation M_s and once the external field is suppressed the ferromagnetic material holds a measurable remanent magnetisation M_r .

Ultimately, ferromagnetism is a phenomenon that cannot be explained solely by classical physics. At the core is the quantum-mechanical concept of *spin exchange coupling*, a phenomenon with no classical counterpart. It was in Landau and Lifshits (1935) that a continuum expression for the exchange energy was obtained. This seminal work and that of W. F. Brown (1940) are the theoretical basis from which micromagnetism emerged. Micromagnetism is a theory that bridges the fundamental quantum-mechanical picture and the effective macroscopic theory of Maxwell equations. Therefore it operates in a scale that is sufficiently large to contain hundreds of atoms making it possible to think of the material as a continuum but not large enough to be described by the effective theory of Maxwell equations. The main goal of a micromagnetic investigation is to obtain a configuration of the magnetic moments in a ferromagnetic material. The main result in Landau and Lifshits (1935) was the theoretical proof that inside

a ferromagnetic material there are regions that are magnetised to saturation called magnetic *domains* and that between these domains exist regions where the magnetisation continuously rotates from the direction of one domain to that of the other, these are called *walls*.

There are two main approaches to micromagnetism. One is obtaining a configuration of the magnetic moments in a ferromagnetic material by minimising the magnetic Gibbs free energy. The other is to solve a partial differential equation that describes the actual dynamics of the magnetic moments; this equation was derived by Landau and Lifshits (1935) and improved by Gilbert, so it's called the Landau-Lifshits-Gilbert equation (LLGE).

2.1.4 Magnetic Gibbs free energy

Chapter 3

Conclusion

3.1 Summary of Thesis Achievements

Summary.

3.2 Applications

Applications.

3.3 Future Work

Future Work.

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