The thermochemical evolution of Mars' deep interior: Geophysical insights with InSight

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NASA's InSight Mission (Banerdt et al., 2012) is due to land on Mars at the end of 2018. It aims to place new constraints on Mars' formation, evolution and present day internal structure. InSight will determine Mars' thermochemical structure using seismic travel times and waveforms (Knapmeyer et al., 2006; Teanby, 2015) recorded by VBB and SP seismometers (the SEIS experiment; Lognonne et al., 2014), by using a heat flow probe (HP3; Spohn et al., 2012) to constrain the thermal gradient in the uppermost lithosphere and by accurately measuring nutations due to the core with an X-band RADAR instrument (RISE; Folkner et al., 2012). These measurements will give us unprecedented constraints on the size and density of the core and on seismic velocities through the planet.

It is imperative that we have the tools and data required to invert the data returned from InSight into constraints on Mars' internal structure, temperature, chemistry and formation. Using a combination of high pressure lab experiments, modelling and seismic data interpretation, I will provide vital data required for analysis of InSight data. **New thermodynamic models** will be created which I will then use to create **realistic seismic models of the Martian interior**. These seismic models will be used to create **synthetic seismograms similar to those expected from the InSight/SEIS experiment**. During the mission, I will conduct **joint geochemical and geophysical inversions from InSight data** to constrain the present-day state and formation history of Mars.

The project will be split into the following work packages:

1. **High pressure experiments to determine the role of iron autoredox reactions under conditions appropriate for Mars' interior.** Despite reducing conditions recorded by the SNC meteorites, high pressures are expected to stabilise Fe^{3+} in silicates in the deep interior, precipitating Fe^0 as a result. This process fundamentally changes the phase relations in the deep mantle and provides a direct link between silicate and sulfide chemistry, yet previous works have not yet incorporated these reactions into models of Mars.

2. **High pressure experiments to isolate the effect of silicate melt composition on high pressure partitioning of trace elements.** One of the primary aims of the InSight Mission is to constrain the chemical composition of Mars' core and conditions of formation. A joint inversion of geophysical data with geochemical data has the potential to provide the strongest constraints, but existing geochemical models are hindered by a poor understanding of the effects of conditions and composition on element partitioning. In this work package, I will conduct multi-anvil experiments to isolate the effect of pressure, temperature and composition on trace element partitioning in silicate melts. The novel experimental design will provide different constraints to those usually provided by partitioning experiments, greatly improving our ability to model core formation and invert data from InSight for the composition of Mars' interior.

3. **Creation of new thermodynamic/thermoelastic models of minerals and melts existing in the deep mantle and core of Mars.** Thermodynamic models are necessary to model seismic velocities in the deep interior as a function of pressure and temperature.

4. **Geophysical modelling of Mars using InSight data**. This work package will involve the vital task of investigating the effects of a comprehensive range of compositional, thermal and structural parameters on seismic waveforms, and trade-offs between them. The results will be vital to maximising scientific return from the mission, especially in the event of limited data. Joint geochemical and geophysical inversions will be designed to provide stronger constraints on the current state and evolution of Mars throughout solar system history.

These additions to our understanding will be used in conjunction with InSight data to fulfil major mission goals, constraining the thermochemical structure of the deep interior and the formation of the planet over 4.5 billion years ago.

(WP1) The ferric iron content and composition of sulfide in Mars' deep mantle

At pressures exceeding ~6 GPa (600 km in Mars), Fe^{2+} in mantle minerals progressively disproportionates, forming Fe^{3+}-bearing silicates (e.g. Frost et al., 2004; Rohrbach et al., 2007) in equilibrium with Fe^0-bearing metal or sulfide phases. These autoredox reactions **fundamentally alter chemical compositions, densities and seismic velocities**, and therefore should be included in planetary models, especially when those planets are iron and sulfide-rich (like Mars). However, there is currently little quantitative data on the extent of these reactions as a function of P, T and chemical composition, without which it is impossible to calculate mineral and melt phase equilibria in Mars' deep interior. In this work package, I will conduct high pressure laboratory experiments to better quantify the effect of pressure and temperature on the redox state of iron **in majoritic garnet under Mars-like conditions**. The results will be combined with the few experimental data from the literature to create a **thermodynamic solution model for garnets**, which can be easily incorporated into software which I will use to calculates densities and seismic velocities as a function of P, T and composition in Mars' deep interior.

Disproportionation of iron in Mars' deep mantle can be described by the following autoredox reaction:

$$3\,Fe_2SiO_4(\text{ringwoodite}) = Fe_3Fe_2Si_3O_{12}(\text{garnet}) + Fe(\text{metal/sulfide}) \qquad \{1\}$$

In the Earth, garnet can accommodate over 25% iron as Fe^{3+} at 14 GPa (Rohrbach et al., 2007; Rohrbach et al., 2011). On Mars, iron contents are much higher than on Earth (Mg# ~ 75 vs. 90; Wanke and Dreibus, 1994), and the mantle is believed to contain markedly more sulfide (~6000 vs <1000 wt ppm FeS; McDonough and Sun, 1995; Tuff et al., 2013). As a result, autoredox is likely to be at least as important on Mars as on Earth, despite highly reducing conditions at the surface. Because of its major impact on the compositions and properties of both silicate and sulfide phases, it is vitally important to quantify the magnitude of the effect.

In this study, I will conduct multi-anvil experiments at high pressure with Mars-like mantle compositions in order to determine the effect of P, T and bulk composition on Fe^{3+} in garnet. Experiments will be run in a 10/4 assembly (Figure 1) at pressures corresponding to Mars' mid-to-lowermost mantle (14, 17 and 20 GPa), and at temperatures of 1600–1900°C. Iron or metal-metal oxides will be used to buffer the iron and oxygen fugacity of the system via the equilibrium (Stagno et al., 2013):

$$2\,Fe_2SiO_4 + 2\,Mg_2SiO_4(\text{ringwoodite}) = Mg_4Si_4O_{12}(\text{garnet}) + 8\,Fe + 4\,O_2 \qquad \{2\}$$

Recovered phases will be analysed for ferric iron content with electron energy loss spectroscopy (EELS).

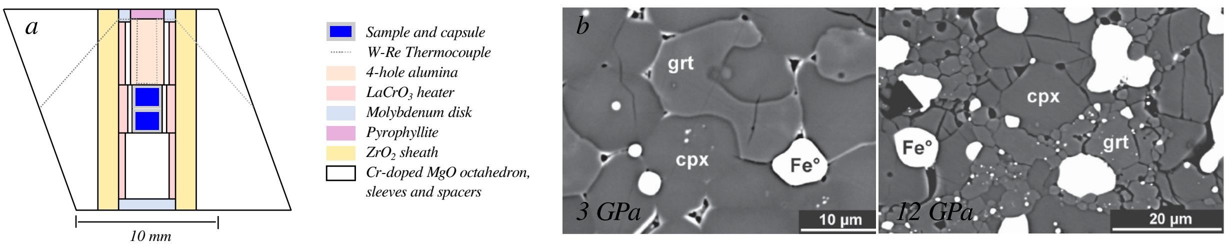

Figure 1: a) Multi-anvil assembly. b) Back-scattered electron images of a recovered charge from multi-anvil runs of Rohrbach et al. (2007), showing precipitation of metallic iron from Fe^{3+}-rich garnet at 12 GPa. Models of Mars' deep interior currently include only ferrous iron, and so cannot accurately model compositions at high pressures.

I will use the results from this work to calculate the amount of ferric iron and composition of sulfide in ultramafic rocks as a function of pressure, temperature and composition (using a model for the effect of sulfur on iron activity). These calculations will be used to create new interior structure models of Mars which include the speciation of iron within mineral and sulfide phases throughout the mantle, and the resultant densities and seismic velocities. One of the exciting things about this project is that the new geophysical data from InSight will reveal the state of major element (dis)equilibrium between Mars' core and mantle.

(WP2) The effect of mantle composition on partitioning of elements during core formation on Mars

The formation of Mars' core was facilitated by large scale melting of the mantle, which allowed large amounts of metallic melt to segregate from the surrounding silicate. Slow diffusion in solid silicates suggests that the compositions of the core and mantle have not changed significantly after formation. Therefore, if there is mantle geochemical data which is sensitive to metal/silicate segregation, it must preserve information relating to core formation. This sensitivity is shown by siderophile and chalcophile elements (elements which are more compatible in metals and sulfides), which are depleted in Mars' mantle (Halliday et al., 2001), and whose "siderophility" is a function of pressure, temperature and composition. As these variables also control the composition of the segregating metal (Rubie et al., 2015), **the mantle abundances of the siderophile elements can be combined with the geophysical data from InSight to constrain the composition of Mars' core**.

High pressure partitioning experiments are typically run for short durations in a reactive MgO capsule and involve complex interactions between Fe-Si-O-C-S metallic melts and multicomponent silicate melts. This means that a) chemical potentials are not held constant during the experiment and b) isolation of the effect of individual model variables is almost impossible. As a result, the results obtained are highly dependent on regression scheme, and have poorly constrained uncertainties (Walter and Cottrell, 2013). Particularly concerning in the case of Mars is the poorly constrained effect of silicate melt composition on high-valence siderophile elements (Righter, 2016). In this work package, I will correct this shortcoming by conducting high pressure experiments on the **solubility of siderophile elements W, Mo, Nb and Ta in silicate melts**. These typically high valence (4^+ to 6^+) elements are **extremely important for determining redox conditions and metal compositions during core formation (O'Neill et al., 2008; Cartier et al., 2014)**.

Multi-anvil experiments will be conducted using **spark-eroded multi-chamber capsules** made out of the element to be studied. Each chamber will be preloaded with a small amount of metal oxide to buffer the system. Glass mixes with **bulk compositions encompassing the range of potential Martian magma ocean compositions** will be loaded into each chamber and then closed. Experiments will be run at **a wide range of pressures and temperatures** (5-20 GPa, 2000-3000 K) until equilibrium is reached (timescales will be assessed during the experiments). 12 samples can be run in a single experiment, effectively **isolating the effect of silicate melt composition**. After the experiment, major and trace element concentrations in the metals, metal oxides and melts will be measured using EPMA and LA-ICP-MS. Activity coefficients $\gamma_{MO_{n/2}}$ will be calculated using the equilibrium reaction between a dissolved metal oxide and its reduced counterpart: $\mathrm{MO_{n/2}}$ (silicate melt) $\rightleftharpoons$ M (metal) + $\frac{n}{4}\mathrm{O}_2$ {3}:

$$\ln \gamma_{MO_{n/2}} = \frac{\Delta G^{\circ}_{\{3\}}}{RT} - \frac{n}{4} \ln f\mathrm{O}_2 + \ln\left(\frac{X_M}{X_{MO_{n/2}}}\right) + \ln \gamma_M \tag{1}$$

The first two terms on the right hand side (functions of the standard state Gibbs energy and oxygen fugacity) are constant at fixed P and T. The third is the partition function $D(M)$, and the fourth is the log of the activity coefficient of M in the metal phase ($\sim$1). I will re-express the composition-dependent $\gamma_{MO_{n/2}}$ values in terms of interaction parameters (e.g., for a Margules model, $A(\mathbf{x}) = \ln \gamma_{MO_{n/2}} / (1 - X_{MO_{n/2}})^2$). Equation 1 can then be used to create accurate models for γ_M in iron-rich metallic melts using data from published iron-metal/silicate experiments. This strategy allows me to investigate the effects of varying P, T and oxygen fugacity on trace element incorporation into silicate melts, which has previously been inaccessible.

I will use the newly created models for trace element incorporation to model the formation of Mars by accretion (e.g. Wade and Wood, 2005) to **predict mantle core compositions and core radius**. I will then **compare these predictions to geophysical data from the InSight Mission** (see WP3; Badro et al., 2015) to constrain the chemical composition of Mars' core. One of the unique benefits of this approach is that it creates **self-consistent multidisciplinary models of Mars' core composition and the conditions under which it formed**.

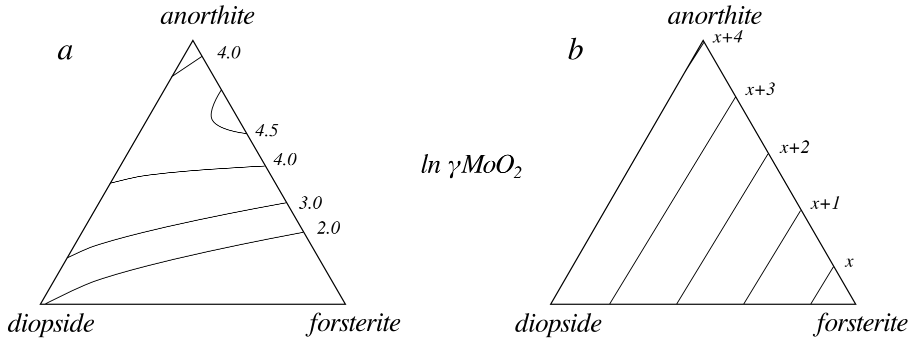

Figure 2: Activity coefficients for MoO_2 at 1650°C, as estimated by a) Wood and Wade (2013) (based partially on 1 bar data in equilibrium with Mo metal) and b) Righter and Chabot (2011) (based solely on high pressure data with iron alloys, subject to an unconstrained shift x). Note the large discrepancies in compositional dependence of γ, despite the fits using much of the same data.

(WP3) Thermodynamic/thermoelastic models of the deep mantle and core of Mars.

Thermodynamic models are a vital requirement of calculating the mineral compositions, density and seismic velocities within Mars as a function of pressure, temperature and composition. Currently, models for high pressure silicates do not include Fe^{3+} (see WP1), and there are no satisfactory thermodynamic models for metallic melts which simultaneously include important elements such as S, Si, O and H. Furthermore, simple models ignore excess properties of solutions, such as bulk moduli, which are likely to play a significant role in determining the properties of metallic solutions (Komabayashi, 2014; Williams et al., 2015). **In this work package, I will create a model for majoritic garnets incorporating ferric iron (see WP1), and add sulfur and hydrogen to a recently-developed self-consistent thermodynamic model for light element incorporation into metallic melts** (Myhill et al., 2016). These additions will allow us to take a potential core composition and **predict densities and seismic properties**, which can then be **compared with those obtained from the InSight Mission**. They will also be able to predict the position of the liquidus for a given composition, **placing bounds on the temperature of Mars' core**.

In creating thermodynamic models it will become possible to self-consistently model chemical equilibrium and seismic velocities. In the case of the majorite model developed from the experimental results of WP1, the changes will allow us to use InSight data to assess the state of equilibrium between Mars' mantle and core. Meanwhile, the new metallic melt model can be interrogated for density and bulk modulus as a function of P, T and composition, enabling the geophysical data from the RISE and SEIS experiments to be inverted for a core composition. Although it is believed that Mars' core is sulfur-rich (Wanke and Dreibus, 1994; Khan and Connolly, 2008), it may contain significant amounts of other elements (Stevenson, 2001), depending on the conditions during core formation. The melt model will also be used to predict depths of an outer-inner core boundary based on core composition and a core-mantle boundary temperature (see WP4), as it can be used to calculate liquid isentropes and liquidi (in conjunction with models of solid metals).

With accurate and comprehensive thermodynamic models, the new data from InSight will be able to answer questions such as the temperature and composition of Mars' mantle, the composition of its core, its evolution and crystallisation regime (Stewart et al., 2007). Depending on the various trade-offs (see WP4), it may be possible to constrain core size without observing core phases, or crystallisation regime of the core even if seismic data cannot unambiguously resolve an inner core.

(WP4) Geophysical modelling of Mars and an assessment of parameter trade-offs and uncertainties.

Creating forward models for Mars' interior provides an efficient way to investigate the effects of different variables on geophysical observables, such as seismic travel times or the moment of inertia. There have not currently been any attempts to systematically investigate the trade-offs between different variables. This is particularly true of Mars' thermal structure and composition, partly because of the unknowns addressed in WP1 and 3, and also partly because of choices of free variables and the richness of information contained in synthetic waveforms. In this work package, **I will build interior models of Mars spanning the full range of plausible thermochemical structures, and analyse the trade-offs and uncertainties in parameters of interest in terms of InSight observables (e.g. P-S differential times, receiver functions, core properties, surface heat flow)**.

Priors for our forward modelling will be the mass, moment of inertia and k2 Love number of Mars, an assumption of chemical equilibrium in the mantle and core and isentropic temperature gradient in the middle of the mantle and within the core. I shall use mineralogical models from the literature (e.g. Stixrude and Lithgow-Bertelloni, 2011), enhanced by the results from WP1,3.

I will investigate the effect of parameters including the Mg number and SiO_2 content of the mantle, thickness of the crust, lithosphere and mantle, composition of the core, and thermal structure parameterised as mantle potential temperature, amount of internal heating and Rayleigh number (a measure of convective vigour). This strategy (which will follow that described in McKenzie et al., 2005) has the benefit of **investigating parameters which are physically meaningful,** with parameters **of geologic interest**, whilst automatically **including often ignored thermal features** such as a lower boundary layer. Recently published thermal models include huge variability in thermal properties; two estimates of the thickness of the conductive lid/thermal boundary layer are 125/∼200 km (Nimmo and Faul, 2013) and 400/∼600 km (Khan and Connolly, 2008). Such variations change the moment of inertia significantly and will therefore affect apparent best-fit core properties. For this reason, **seismic waveforms recorded by the SEIS experiment should provide important constraints on core properties, even if no core phases are observed.**

I will use the forward models which fit the prior constraints (within uncertainties) to create **synthetic waveforms (using TauP, SpecFem)**. I am currently collaborating with Stefanie Hempel (ISAE Toulouse) to set up the required workflow, taking advantages of new software functionality such as the ability to create synthetics for an oblate spheroid. From these, I will identify diagnostic waveform properties and parameter trade-offs. This study will **enable rapid interrogation of data when it arrives from InSight**.

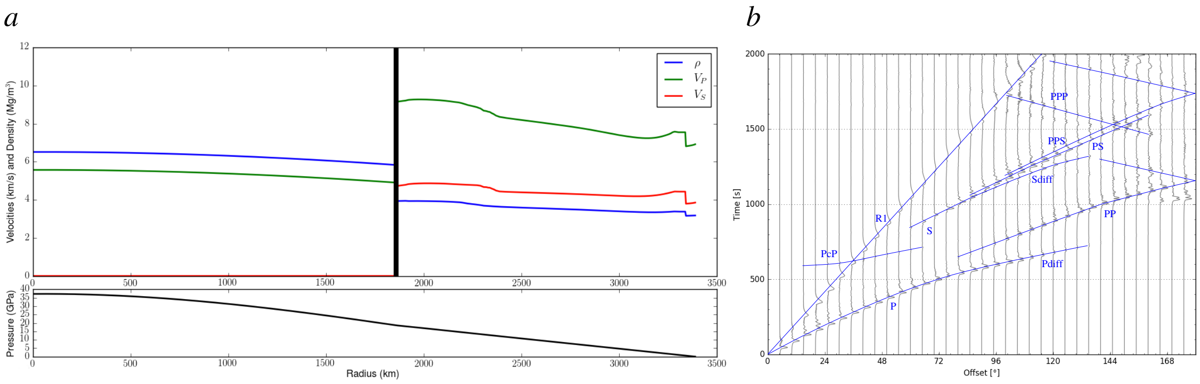

Figure 3: a) Example 1D seismic model of Mars, fitting the mass and moment of inertia of the planet and geochemical estimates of heat-producing elements (U, Th and K). Model includes thermal boundary layers at the top and bottom of the convecting mantle, and a mechanical boundary layer with experimentally-constrained temperature-dependent conductivity. b) Example record section created from a 1D Mars seismic model. Sections like these will be interrogated for seismic arrival times/amplitudes and waveshapes to provide a set of diagnostic tools for the InSight Mission.

Monitoring of progress

My intended research progress during the project can be found in Table 1. A fully detailed plan will be delivered to the university on starting the fellowship. Annual staff reviews are routinely carried out with a senior member of the research staff to ensure research goals are met.

Any problems with experimental apparatus at the University of Bristol can be remedied via use of facilities in BGI, Germany, but it is expected that the vast majority of work will be performed in-house. In terms of personal skills, Bristol University runs staff development courses on a wide variety of subjects including project management and research leadership.

Table 1: Proposed workflow. E=Experiments, A=Analysis, M=Modelling, Wx=write-up of paper x

Term starting	01/18	05/18	09/18	01/19	05/19	09/19	01/20	05/20	09/20
WP1 (Majorite)	E	A	W1	W2					
WP2 (Partitioning)			E	E	A	W3	A	W4	
WP3 (Modelling)		M	M/W5			M	W6		
WP4 (Mars Structure)				A	A/W?	A/W?	A/W?	A/W?	A/W?

Research Environment

The School of Earth Sciences at the University of Bristol has an ideal research environment for this project. It houses two multi-anvil presses and an array of analytical apparatus which is available and can be used to fulfil the experimental parts of this project. An in-house linux cluster and University facility BlueCrystal are available for high performance computing. Permanent staff include Nick Teanby and James Wookey, who are both members of the InSight Team, and with whom I am currently working as a PDRA on a UKSA grant to study InSight seismometer deployment, regolith properties, and shallow seismic events. During the experimental part of this project, I shall be working with Professor Michael Walter, known for his high-pressure experimental work related to deep planetary interiors. The School of Earth Sciences has over 70 PhD students and some 50 Masters students.

During this project, I will also spend some time at the Bayerisches Geoinstitut, where I previously spend three years as a Humboldt Research Fellow. The Bayerisches Geoinstitut is a specialist high pressure experimental petrology institute attached the the University of Bayreuth, Germany. It has a wide variety of experimental and analytical apparatus. At any one time it has 15-20 PhD students and 5-10 Masters students.

Justification for resources requested

Salaried Costs: I will spend 100% of my time on the project, the salary is in line with standard UoB banding for a research fellow with 5 years of experience.

Computing Costs: The main computing requirement is a desktop that is sufficiently high-specification to create synthetic waveforms from high resolution interior structure models using SpecFEM (3500 GBP). Other costs include a laptop for lab and conference work (1000 GBP), data archival hardware (1500 GBP) and other lab and computing consumables (1000 GBP).

Experimental Costs: Most multi-anvil experiments (WP1: 3x3 experiments covered three pressure and temperature points; WP2: 4x3x3 experiments covering four elements, three pressure and temperature points) will be conducted at the University of Bristol, with some large volume/high pressure runs at the Bayerisches Geoinstitut, Germany. It is expected that there will be some heater failures due to $LaCrO_3$ imperfections. For this reason, five extra experiments are included in the costs. Costs per multianvil experiment take into account capsule and starting materials, assembly costs ($\sim$ 50 GBP), cube breakage ($\sim$200 GBP/cube), maintenance and machining, and analysis (LA-ICP-MS, majors via EPMA and ferric iron with EELS).

Travel costs: During the two years of the mission, there will be quarterly team meetings, but as a relatively junior member of the team, I shall not be required to attend all meetings. Two per year should be sufficient. On top of this, I request funds to present results at one conference per annum - I intend to present results from this project at the LPSC (Texas) and the AGU Fall Meeting (normally in San Francisco). Finally, as I shall be working with researchers in Germany (Professor Dan Frost, BGI) and France (Dr Stefanie Hempel, ISAE Toulouse), I request funds for one collaborative visit per annum.

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