# Plausibility of lunar crustal magnatism producing strong crustal magnetism

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#### Abstract

The Moon generated a long-lived core dynamo magnetic field, with intensities at least episodically reaching ~10–100  $\mu$ T during the period prior to ~3.56 Ga. While magnetic anomalies observed within impact basins are likely attributable to the presence of impactor-added metal, other anomalies such as those associated with lunar swirls are not as conclusively linked to exogenic materials. This has led to the hypothesis that some anomalies may be related to magmatic features such as dikes, sills, and laccoliths. However, basalts returned from the Apollo missions are magnetized too weakly to produce the required magnetization intensities (>0.5 A/m). Here we test the hypothesis that subsolidus reduction of ilmenite within or adjacent to slowly cooled mafic intrusive bodies could locally enhance metallic FeNi contents within the lunar crust. We find that reduction within hypabyssal dikes with high-Ti or low-Ti mare basalt compositions can produce sufficient FeNi grains to carry the minimum >0.5 A/m magnetization intensity inferred for swirls, especially if ambient fields are >10  $\mu$ T or if fine-grained Fe-Ni metals in the pseudo-single domain grain size range are formed. Therefore, it is plausible that the magnetic sources responsible for long sublinear swirls like Reiner Gamma and Airy may be magmatic in origin. Our study highlights that the domain state of the magnetic carriers is an under-appreciated factor in controlling a rock's magnetization intensity. The results of this study will help guide interpretations of lunar crustal field data acquired by future rovers that will traverse lunar magnetic anomalies.

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# **1** Plausibility of lunar crustal magmatism producing strong crustal magnetism

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# 7 Key Points:

- 8 Ilmenite reduction experiments were run at a wide range of experimental conditions
- Subsolidus reduction of ilmenite at lunar conditions creates magnetizable products
- Swirl magnetic source bodies may be caused by the cooling of high-Ti basaltic dikes

# 11 Abstract

12 The Moon generated a long-lived core dynamo magnetic field, with intensities at least 13 episodically reaching  $\sim 10-100 \mu T$  during the period prior to  $\sim 3.56$  Ga. While magnetic 14 anomalies observed within impact basins are likely attributable to the presence of impactor-15 added metal, other anomalies such as those associated with lunar swirls are not as conclusively 16 linked to exogenic materials. This has led to the hypothesis that some anomalies may be related 17 to magmatic features such as dikes, sills, and laccoliths. However, basalts returned from the 18 Apollo missions are magnetized too weakly to produce the required magnetization intensities 19 (>0.5 A/m). Here we test the hypothesis that subsolidus reduction of ilmenite within or adjacent 20 to slowly cooled mafic intrusive bodies could locally enhance metallic FeNi contents within the 21 lunar crust. We find that reduction within hypabyssal dikes with high-Ti or low-Ti mare basalt 22 compositions can produce sufficient FeNi grains to carry the minimum >0.5 A/m magnetization 23 intensity inferred for swirls, especially if ambient fields are >10  $\mu$ T or if fine-grained Fe-Ni 24 metals in the pseudo-single domain grain size range are formed. Therefore, it is plausible that 25 the magnetic sources responsible for long sublinear swirls like Reiner Gamma and Airy may be 26 magmatic in origin. Our study highlights that the domain state of the magnetic carriers is an 27 under-appreciated factor in controlling a rock's magnetization intensity. The results of this study 28 will help guide interpretations of lunar crustal field data acquired by future rovers that will 29 traverse lunar magnetic anomalies.

30 Plain Language Summary

While the Moon does not have a magnetic field today, some parts of its crust such as impact basins and bright and sinuous features called "lunar swirls" are still magnetized. Strongly magnetized regions observed within impact basins could be related to iron-rich material derived

34 from impactors. However, other magnetized regions, such as those associated with lunar swirls, 35 are not as conclusively linked to externally added materials. It has been proposed that the 36 strongly magnetic regions associated with lunar swirls are related to lunar igneous intrusive 37 rocks. Here we experimentally test the hypothesis that the thermal alteration of FeTiO3 grains to 38 TiO2 grains and metallic iron within or next to slowly cooled igneous intrusive features while 39 the Moon had a magnetic field, could explain the strong magnetic regions associated with lunar 40 swirl. We show that the lunar swirl minimum magnetization intensity can be reached from the 41 thermal alteration of ilmenite, especially if ambient fields are strong enough or if fine-grained 42 Fe-Ni metals are formed. This study will help interpret data acquired by future rovers traversing 43 magnetic anomalies on the lunar surface.

### 44 **1 Introduction**

45 Paleomagnetic studies have suggested that the Moon may have generated a core dynamo magnetic field at least intermittently between ~4.25 Ga and ~1.5 Ga, with intensities reaching 46 47 ~40–110 µT prior to ~3.56 Ga (Weiss and Tikoo, 2014; Tikoo and Evans, 2022; Wieczorek et al., 48 2022). The absence of magnetization within young lunar breccias suggests that the dynamo 49 likely ceased between 1.92 and 0.80 Ga ago (Mighani et al., 2020). Whether the dynamo 50 operated continuously and exactly when the lunar dynamo ceased remain uncertain (Evans et al., 51 2018; Tarduno et al., 2021). The dynamo history of the Moon is also evident from its remanent 52 crustal magnetism (Hood, 2011; Hood et al., 2021; Purucker et al., 2012; Wieczorek et al., 2022). 53 Intense magnetic anomalies within impact basins are likely caused by impactor-added metal 54 within melt sheets (Oliveira et al., 2017), but anomalies associated with lunar swirls such as the archetypal Reiner Gamma (Denevi et al., 2016; D. Hemingway & Garrick-Bethell, 2012) and 55

Airy albedo features (David T. Blewett et al., 2011) are more difficult to unequivocally attribute
to exogenic metal.

58 The bulk of remanent magnetization on the Moon is likely recorded within grains of 59 metallic iron and iron-nickel alloys within crustal and upper mantle rocks (Weiss & Tikoo, 2014; 60 Wieczorek, 2018). Anomalies on the southern lunar farside have variably been hypothesized to 61 be related to metal-rich ejecta from the South Pole-Aitken impactor (Wieczorek et al., 2012) or 62 strongly magnetized mafic dikes (Purucker et al., 2012). However, the origin of magnetization at 63 lunar swirls is even more enigmatic because swirls frequently lack correlations with distinct 64 geological features (Denevi et al., 2016). It is hypothesized that swirl-affiliated magnetic 65 anomalies could be related to buried impact melt sheets, impactor-derived ejecta, and iron-rich dikes (Garrick-Bethell & Kelley, 2019; D. J. Hemingway & Tikoo, 2018). However, basalts 66 67 returned from Apollo missions are generally weakly magnetized (<0.01 A/m) (Wieczorek et al., 68 2012) and are incapable of producing the magnetization intensities required for swirl formation 69 (>0.5 A/m) (D. J. Hemingway & Tikoo, 2018). It was recently proposed that subsolidus 70 reduction of ilmenite and other minerals within or adjacent to mafic dikes that cooled very 71 slowly (over thousands of years depending on dike width) could produce elevated iron metal 72 contents within the lunar crust (D. J. Hemingway & Tikoo, 2018). Following the approach of 73 Oliveira et al. (2017), Hemingway and Tikoo (2018) determined that reaching magnetization 74 intensities of >0.5 A/m (assuming a 1-km magnetic source thickness) requires rocks to 75 contain >0.3 wt. % Fe-metal. Mare basalts typically have lower metal contents of ~0.08 wt. % Fe 76 (Gose & Butler, 1975). However, Hemingway and Tikoo (2018) calculated that in the extreme 77 case where all ilmenite in a mare basalt were to be converted to  $Fe + TiO_2$ , rocks of basaltic 78 composition could be enriched by up to 11 wt.% Fe. Due to uncertainties regarding the rates of

79	thermochemical alteration of ilmenite, it is unclear whether subsolidus reduction could produce
80	iron concentrations high enough to be consistent with anomalies present at swirls. In this study,
81	we experimentally test the hypothesis that subsolidus reduction of ilmenite can enhance metallic
82	FeNi contents within the lunar crust when it occurs in or near cooled mafic intrusive bodies.
83	
84	2 Experimental and Analytical procedures
85	2.1 Starting Material
86	We used a kimberlitic ilmenite megacryst from Kumgbo, Liberia as the starting material.
87	These natural samples were described in Haggerty (2017), and there were two classes of ilmenite:
88	high-Mg and low-Mg class. The sample we were using for starting material was a high-Mg end
89	member. The high-Mg ilmenite had the lowest amount of ferric iron and was a better match for
90	lunar ilmenites which are Mg-rich (up to 6 wt.%) and ferric-iron-poor (Mason & Melson, 1970).
01	



92

93 Fig. 1: a. a representative backscatter electron image of a 1500 X view on ilmenite starting material. b. Backscatter electron image of a 110 X view of experimental run H182\_S ( $fO_2 =$ 94 95 IW-1 cooling experiment from 800-500 °C by 3 °C/hour) showing both homogeneous nonreacted middle area and reacted rim. c. Backscatter electron image of experimental run H171 96 97  $(fO_2 = IW-0.5 \text{ cooling experiment from 800-500 °C by 3 °C/hour)}$ . The dendritic high 98 reflectance material is FeNi metal formed via subsolidus reduction of ilmenite, and the metal 99 blebs were formed along with the rutile within the reaction zone. Yellow lines separated the 100 light grey unreacted starting material zone from the dark grey reaction zone. A linear traverse of electron microprobe analyses crossing non-reacted to reacted zones was taken to study the 101 102 compositional difference (blue line). d. Backscatter electron image of experimental run H158 ( $fO_2 = IW-2$  isothermal experiment run at 800 °C). The colors were the same as in c. 103

- 104
- 105 2.2 Experimental Setup

106 Ilmenite reduction experiments were performed in one-atmosphere gas-mixing vertical 107 tube furnaces at a wide range of durations, quenching temperatures, and grain size of starting

108 material. Samples were suspended in Re baskets. The Re-wire basket is then attached to a small-109 diameter Pt quench wire hanging from two thicker Pt electrodes as part of a sample holder 110 assembly. Before the sample holder was inserted into the furnace, the temperature and  $fO_2$  of the 111 system were set at the desired experimental conditions. The temperature during an experiment 112 was monitored by two R-type thermocouples that were calibrated against the melting point of gold and had an estimated accuracy of  $\pm 2^{\circ}$ C. Oxygen fugacity was controlled using an H<sub>2</sub>-CO<sub>2</sub> 113 114 gas mixture in all experiments. The  $fO_2$  of each experiment was monitored in situ using a yttria-115 doped oxygen sensor from Ceramic Oxide Fabricators, with an estimated accuracy of ±0.05 116  $\log fO_2$  units. Three batches of experiments were performed: 1) time series of isothermal 117 reduction experiments at constant  $fO_2$ , 2) cooling series at constant  $fO_2$ , and 3) a series with 118 constant  $fO_2$  and cooling histories but differing ilmenite grain sizes (Table 1). For the first batch 119 of experiments, all experiments were conducted at 800 °C and  $fO_2 = IW-1$  with different time 120 duration (2 days, 4 days, 8 days, and 16 days). Ilmenite grain sizes for the first batch of 121 experiments ranged between 1-3 mm. For the second batch of cooling experiments, we used 122 multiple cooling paths. One experiment involved heating ilmenite at 800 °C for 48 hours and 123 then cooling it to 500 °C at a rate of 3 °C/hour ( $fO_2 = IW-0.5$ ) (experimental run H171), while a 124 second experiment in this batch involved heating at 1000 °C for 48 hours and then cooling to 500 125 <sup>o</sup>C at a rate of 3 <sup>o</sup>C/hour ( $fO_2 = IW-1$ ) (experimental run H174). The  $fO_2$  sensor was removed 126 before the temperature dropped below 800 °C for the cooling experiments to protect the fO<sub>2</sub> 127 sensor. The  $fO_2$  sensor was only rated for below 1200 mV and decreasing temperature would 128 increase the absolute mV values to greater than 1200 mV. In the third batch of experiments 129 ilmenite grain sizes were systematically varied. In addition to 1-3 mm ilmenite chips, we also

- included 10–15 0.5 mm diameter ilmenite pieces in these two experiments. For the third batch of
  experiments with different grain sizes of ilmenite
- 132 (experimental runs H181 and H182), all experiments were heated at 800 °C for 48 hours and then
- 133 cooled to 500 °C at 3 °C/hour ( $fO_2 = IW-1$ ). The  $fO_2$  sensor was removed before the
- 134 temperature dropped below 800 °C during these experiments as well. Since the degree of
- 135 subsolidus reduction may depend on the exposed surface area to volume ratio of a given ilmenite
- 136 grain, the starting materials for each set of experiments were divided into small, medium, and
- 137 large to study the relationship between starting materials' grain sizes and magnetization
- 138 properties. Experimental runs H181\_S and H182\_S involved ilmenite grain sizes ranging
- between 0.5–1.0 mm in diameter. Ilmenite grain sizes in runs H181\_M and H181\_M were
- 140 between 1.2–1.6 mm in diameter. Finally, starting materials grain sizes in runs H181\_L and
- 141 H181\_L ranged between ~2.0–4.0 mm in diameter. Table 1 summarized the run conditions
- 142 including the initial temperature, duration, cooling rate, grain size, oxygen fugacity, and
- 143 experimental temperatures for all experiments.

Exp	Temp (°C)	fO <sub>2</sub>	Duration (days)	Ilmenite Diameter (mm)	$M_{\rm s}$ (Am <sup>2</sup> /kg)	$M_{\rm rs}$ (Am <sup>2</sup> /kg)	$M_{ m rs}/M_{ m s}$	<i>B</i> <sub>c</sub> (mT)	B <sub>cr</sub> (mT)	$B_{\rm cr}/B_{\rm c}$	Geik	Ilm	Hem	Hysteresis Mass (mg)
S.M.				~1–3	n /a	0.00	n /a	0.00	n /a	n /a	48.65	48.77	2.58	34
H155	800	IW-1	16	~1–3	3.43 x 10 <sup>-2</sup>	1.65 x 10 <sup>-3</sup>	4.81 x 10 <sup>-2</sup>	16.90	174.00	10.30	52.41	45.79	1.80	14
H154	800	IW-1	4	~1–3	3.20 x 10 <sup>-2</sup>	2.50 x 10 <sup>-3</sup>	7.80 x 10 <sup>-2</sup>	17.70	75.00	4.24				21
H158	800	IW-2	4	~1–3	1.94	5.24 x 10 <sup>-2</sup>	2.70 x 10 <sup>-2</sup>	2.30	22.00	9.57	57.17	43.10	0.00	28
J137	800	IW-1	8	~1–3	7.31 x 10 <sup>-3</sup>	8.23 x 10 <sup>-4</sup>	1.13 x 10 <sup>-1</sup>	18.40	62.00	3.37	50.95	49.44	0.00	32
J138	800	IW-1	2	~1–3	5.50 x 10 <sup>-3</sup>	1.17 x 10 <sup>-3</sup>	2.13 x 10 <sup>-1</sup>	27.10	300.00	11.07	50.33	46.32	3.35	17
H171	800–500	IW- 0.5	6.2	~1–3 (3), ~0.5 (10)	8.13	6.50 x 10 <sup>-1</sup>	8.01 x 10 <sup>-2</sup>	4.10	10.00	2.44	78.43	21.00	0.57	9
H174	1000–500	IW-1	8.9	~1–3 (3), ~0.5	4.13 x 10 <sup>-1</sup>	1.20 x 10 <sup>-2</sup>	2.90 x 10 <sup>-2</sup>	4.80	47.00	9.79	51.15	48.43	0.41	10
H181_S	800-500	IW-1	6.2	0.86-0.47	4.78 x 10 <sup>-1</sup>	8.55 x 10 <sup>-2</sup>	1.79 x 10 <sup>-1</sup>	13.70	46.30	3.38	64.35	33.08	2.57	31
H181_M	800-500	IW-1	6.2	1.37–1.11	2.77 x 10 <sup>-1</sup>	4.91 x 10 <sup>-2</sup>	1.77 x 10 <sup>-1</sup>	16.30	53.20	3.26				31
H181_L	800-500	IW-1	6.2	3.97-1.96	2.40 x 10 <sup>-1</sup>	4.50 x 10 <sup>-2</sup>	1.88 x 10 <sup>-1</sup>	8.70	33.60	3.86				31
H182_S	800–500	IW-1	6.2	1.09-0.52	1.41	1.33 x 10 <sup>-1</sup>	9.43 x 10 <sup>-2</sup>	6.00	14.80	2.47	62.11	34.22	3.66	19
H182_M	800-500	IW-1	6.2	1.61–1.23	8.41 x 10 <sup>-1</sup>	8.15 x 10 <sup>-2</sup>	9.69 x 10 <sup>-2</sup>	5.30	27.00	5.09	62.21	34.78	3.01	31
H182_L	800–500	IW-1	6.2	3.97–1.96	6.66 x 10 <sup>-1</sup>	6.08 x 10 <sup>-2</sup>	9.13 x 10 <sup>-2</sup>	4.80	16.60	3.46				31

Table 1: Run conditions and hysteresis analyses of starting materials and experimental run products

Note: S.M. stands for starting material. Experimental runs H171 and H174 were held at 800 °C and 1000 °C respectively for 48 hours then cooled to 500 °C by 3 °C/hour; experimental runs H181 and H182 were held at 800 °C for 48 hours and then cooled by 3 °C/hour to 500 °C. Geikielite, ilmenite, and hematite compositions were approximated because they were calculated from EPMA spot analyses. See Supplementary Materials on how Geik, Ilm, and Hem compositions were calculated.

152 Rock magnetic experiments (magnetic hysteresis and backfield remanence) were 153 performed on ilmenite starting material as well as reduced products using a LakeShore 8600 154 Vibrating Sample Magnetometer (VSM) instrument at the Institute for Rock Magnetism at the 155 University of Minnesota. These experiments elucidate the grain size of magnetic minerals and 156 the magnetization carrying capacity of a sample. During a magnetic hysteresis experiment, a 157 sample was placed within a VSM in an initially zero field. The field (B) was increased to an 158 intensity of +1 Tesla (T) in the positive direction, before being reduced in intensity and then 159 applied in the reverse direction to the same intensity (i.e., -1 T) prior to cycling the field back up 160 to +1 T. During the hysteresis experiment, the magnetization intensity of the sample (M) was 161 continuously measured. Magnetic hysteresis properties were obtained following a slope 162 correction to remove paramagnetic contributions.  $M_s$  is the saturation magnetization (the 163 strongest magnetization the ferromagnetic component of a sample can contain, in the presence of 164 a saturating field).  $M_{\rm rs}$  is the saturation remanent magnetization (the residual remanence after a 165 saturating field is removed).  $B_c$  is the coercive field (a measure of the ability to withstand an 166 external field without becoming remagnetized). During backfield experiments, a sample is 167 imparted with a saturating +1 T magnetization in one direction. Then direct fields are applied in 168 the opposite orientation with increasing magnitude to -1 T.  $B_{cr}$  is the coercivity of remanence (a 169 measure of the required magnetic field to null an initial saturation remanent magnetization 170 acquired from a field with opposite orientation) (Day et al., 1977). Here we used  $M_{\rm rs}$  to help 171 quantify the remanence-carrying ability of ferromagnetic material produced during our 172 subsolidus reduction experiments. Hysteresis and backfield data were processed using the 173 HystLab software package (version 1.0.10) written for MATLAB (Paterson et al., 2018). Values 174 of  $M_{\rm s}$ ,  $M_{\rm rs}$ ,  $B_{\rm c}$ , and  $B_{\rm cr}$  for each experiment were shown in Table 1.

175 2.3 Post-experimental material characterization

176 After the reduction experiments, samples were studied by an electron microprobe and a 177 petrographic microscope. Concentrations of major elements in experimental products were 178 analyzed using a JEOL JXA-8200 electron microprobe equipped with five wavelength-dispersive 179 spectrometers, and a JEOL (e2v / Gresham) silicon-drift energy-dispersive spectrometer at 180 Washington University in St. Louis. Analyses were acquired using Probe for EPMA software. 181 A focused beam (~1 µm in diameter) with a current of 25 nA at a 15 kV accelerating voltage was 182 used to measure the concentrations of oxides and Fe-Ni metals. Background X-ray corrections 183 were performed on glass using the mean atomic number correction (Carpenter, 2016).

184

### 185 **3 Results**

186

#### 3.1 Ilmenite subsolidus reaction products

187 The ilmenite megacryst appeared homogeneous before reduction (a characteristic BSE 188 image of the sample is shown in Fig. 1a). The composition of the starting material was on 189 average 54.98 wt.% Geikielite, 40.56 wt.% ilmenite, and 4.46 wt.% hematite. EPMA analyses of 190 starting materials and experimental run products were reported in Table 2. After reduction 191 experiments, there were reaction zones along the edges of the sample chips and along pre-192 existing cracks. The reduced megacryst material was darker than the non-reacted starting 193 material area in BSE images indicating it is more Mg-rich, and the reaction rims are ~10-50 µm 194 wide (Fig. 1b-d). Reaction products consisted of pure ilmenite (i.e., no hematite solid solution), 195 Cr-spinel, rutile exsolution, and 1–10  $\mu$ m diameter nodules of kamacite ( $\alpha$ -Fe<sub>1-x</sub>Ni<sub>x</sub> for x < ~0.05) 196 (Fig. 1c and 1d, Table 3). For the experiment run at  $fO_2 = IW-0.5$  and had a cooling history of 197 800–500 °C (experimental run H171), the composition of the reaction zone changed from

198	Geik48Ilm49Hem3 of the starting material to Geik78Ilm21Hem1. There were 1–10 µm metals
199	present in the reaction zone, and the average composition of metal was Fe99Ni1 (kamacite).
200	Rutile veins were visible within reaction zones (Fig. 1c). The presence of rutile was confirmed
201	by EPMA and had a composition of pure $TiO_2$ (Fig. 2a). The composition of the reaction zone
202	changed from Geik48Ilm49Hem3 of the starting material to Geik57Ilm43 for an experiment run
203	at $fO_2 = IW-2$ and 800 °C (Experimental run H158). BSE images reveal abundant 1–3 µm metal
204	grains found in the reaction zones (Fig. 1d). The average composition of the metal was Fe98Ni2
205	(kamacite).

	No.	SiO		TiO		$Al_2$		Cr <sub>2</sub>		Fe		Mn		Mg		Ca		Ni		Zn		$Na_2$		$K_2$	
	of analy	2		2		O <sub>3</sub>		O <sub>3</sub>		0		0		0		0		0		0		0		0	
	ses																								
S.M.	41	0.0	0.0	55.	0.2	0.65	0.0	1.92	0.0	27.	0.2	0.2	0.0	14.	0.0	0.0	0.0	0.2	0.0	0.0	0.0				
		0	8	02	6		7		3	55	7	6	2	37	8	3	4	1	2	2	2				
H155	4	0.0	0.1	56.	1.0	0.58	0.2	2.03	0.6	25.	1.0	0.2	0.0	14.	0.2	0.0	0.0	0.7	1.1	0.0	0.0				
		7	8	12	5		1		0	83	8	6	2	44	3	4	1	5	7	4	2				
H158	10	0.0	0.0	58.	1.6	0.81	0.5	2.07	1.2	22.	0.4	0.3	0.0	16.	0.1	0.0	0.0	0.0	0.0	0.0	0.0				
		0	7	37	3		4		7	09	1	6	5	97	3	7	3	4	4	1	3				
J137	3	0.0	0.0	57.	1.8	0.48	0.4	1.83	1.2	25.	0.1	0.2	0.0	14.	0.2	0.0	0.0	0.1	0.0	0.0	0.0				
		0	1	42	9		2		9	42	7	7	2	98	3	3	1	2	1	1	2				
J138	4	0.0	0.0	55.	0.1	0.80	0.1	1.94	0.0	27.	0.3	0.2	0.0	15.	0.2	0.0	0.0	0.2	0.0	0.0	0.0				
		3	5	45	4		7		2	96	3	6	1	02	6	3	2	2	2	4	1				
H171	6	0.0	0.0	61.	0.7	0.50	0.1	1.95	0.3	11.	0.7	0.4	0.0	23.	0.6	0.0	0.0	0.0	0.0	0.0	0.0	0.02	0.0	0.0	0.0
		0	2	37	0		7		8	96	6	8	3	84	7	2	1	3	1	3	3		1	0	0
H174	4	0.0	0.0	58.	0.2	0.19	0.0	0.73	0.1	25.	0.2	0.2	0.0	14.	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.05	0.0	0.0	0.0
		0	2	43	6		9		6	57	1	9	4	85	5	3	1	6	7	2	3		3	1	1
H181_	3	0.0	0.0	58.	0.2	0.51	0.1	1.63	0.5	20.	0.4	0.3	0.0	19.	1.0	0.0	0.0	0.0	0.0	-	0.0	0.03	0.0	0.0	0.0
S		0	8	82	0		1		8	10	9	1	3	01	0	2	1	0	1	0.0 1	2		3	0	1
H182_	9	0.0	0.0	57.	0.7	0.87	0.5	2.19	0.7	20.	0.9	0.3	0.0	18.	0.5	0.0	0.0	0.0	0.0	0.0	0.0	0.01	0.0	0.0	0.0
S		0	3	71	5		1		0	57	0	1	3	18	6	1	1	2	1	3	3		2	0	0
H182_	2	0.1	0.0	56.	0.6	1.04	0.3	3.18	1.2	20.	0.0	0.2	0.0	18.	0.7	0.0	0.0	0.0	0.0	0.0	0.0	0.05	0.0	0.0	0.0
Μ		0	5	49	9		5		9	99	9	9	0	84	9	1	0	4	2	5	0		1	0	0

#### · · • • • 1 4 200

experiments, we reported the average compositions and their one standard deviation of the top 10% highest geikielite components of 208

the total analyses of each experimental run. The analyses in this table were taken from the reaction zone on the grains. Columns on 209

210 the right of each oxide were each oxide's one standard deviation from the number of analyses for each experimental run. The full

211 EPM spot analyses of the starting materials and each experimental run can be found in the Supplementary Materials. Experimental

runs H154, H181\_M, H181\_L, and H182\_L were not analyzed by the EPMA, but their hysteresis analyses were reported in Table 1. 212

213 Table 3: Metal compositions (at.%) for experiments that formed  $> 5 \mu m$  metals 214

	No. of analyses	Fe*	Ni*
H158	12	97.99	2.01
H171	5	99.28	0.72
H182_M	7	98.38	1.62
H182_L	1	98.59	1.41

- 215 Note: \*These compositions were approximated because they were calculated from EPMA spot
- analyses that overlapped with ilmenite. The FeNi metals were all close to the bcc regime and not 216
- close to the fcc regime. See Supplementary Materials on how Fe and Ni compositions were 217 calculated.
- 218
- 219





221 222

223 Fig. 2: Scattered plots of the Ti APFU for balanced stoichiometric ilmenite vs. Mg APFU 224 balanced stoichiometric ilmenite and geikielite solid solution of reduction experimental runs 225 H171 (a) and H158 (b) (blue dots) comparing to the starting materials (orange dots). Each dot represented one EPMA data point. Experimental data points (blue) compiled all EPMA traverses 226 227 from each experiment. Experimental run H171 traverse EPMA data points showed a mixing 228 feature between two end members: rutile and ilmenite+geikielite solid solution. "Mixing line" in 229 Fig. 2a refers to EPMA points that overlapped with/or contained small rutile crystals. Rutile 230 recalculated to ilmenite is Ti<sub>1.5</sub>O<sub>3</sub> with 1.5 APFU Ti. A comparison of the backscattered electron 231 images of runs H171 and H158 can be seen in Fig. 1c and Fig. 1d.

232

We did traverse analyses by EPMA across the ilmenite reduction reaction zones and found that they have less hematite compared to the starting material. There was an increasing trend of the Ti formula units for balanced stoichiometric ilmenite for all reduced experiments

16

(Fig. 2). Mg formula units for balanced stoichiometric ilmenite and geikielite solidus solution
trending showed both excess and deficit for reduced experiments. Experimental runs H158 and
H17 had both increasing Ti and Mg formula patterns (Fig. 2).

Our magnetic hysteresis experiments indicated that the starting material was paramagnetic, as expected for ilmenite at room temperature ( $M_{rs} = 0$ ). In contrast, all subsolidus reduction products contained ferromagnetic material, as evidenced by their hysteresis loops and parameter values (Figure S1, Table 1). Based on the hysteresis parameter values ( $M_{rs}/M_s < 0.5$ and  $B_{cr}/B_c > 1.5$ ), the FeNi grains in the reduced samples were likely in the pseudo-single domain (PSD) to multidomain (MD) size range (Day et al., 1977).

245

246

3.2 Reduction isothermal experiments (800 °C) time series at constant  $fO_2$ 

The reduced experiments run for different experimental durations had  $M_{\rm rs}$  values of 247  $5.50*10^{-3}$  Am<sup>2</sup>/kg (experimental run J138 for 2 - day),  $2.50*10^{-3}$  Am<sup>2</sup>/kg (experimental run H154 248 for 4 - day),  $8.23*10^{-4}$  Am<sup>2</sup>/kg (experimental run J137 for 8 - day), and  $1.65*10^{-3}$  Am<sup>2</sup>/kg 249 (experimental run H155 for 16 - day). Following our initial IW-1 experiments,  $M_{rs}$  values 250 251 increased substantially from the zero value of the starting material, indicating that kamacite was 252 likely the created phase rather than taenite (the latter is paramagnetic at room temperature for 253 <30% Ni). Within this batch of experiments, there was no obvious correlation between the  $M_{\rm rs}$ 254 values and the experimental durations (Fig. 3). We posit that the observed variations in  $M_{\rm rs}$  were 255 most likely dominated by the nonuniform density of random internal fractures inherent in the 256 starting material that can differ between subsamples.



257

258

Fig. 3: Time series of isothermal reduction experiments. The comparison of isothermal experiments of different time durations at  $fO_2 = IW-1$  and the starting material. Black dot represented the  $M_{rs}$  value for the starting material, and blue dots represented  $M_{rs}$  values for the isothermal reduction experiments. All experiments have larger  $M_{rs}$  than starting materials, but no trend with time. Suggesting surface area is a dominant factor.



# 3.3 Reduction experiments with slow cooling

To study the actual cooling process of ilmenite in the Moon's crust, we did two cooling experiments from 800–500 °C at  $fO_2 = IW-0.5$  (experimental run H171) and 1000–500 °C at  $fO_2$ = IW-1 (experimental run H174). Despite the uncertainties on the  $fO_2$  conditions (as this run was initially intended to take place at IW-1), H171 had  $M_{rs}$  values at least 1 order of magnitude higher than the other isothermal experiments, and H174 had  $M_{rs}$  values similar to the other isothermal experiments (Table 1). The origin of the high  $M_{rs}$  value of the  $fO_2 = IW-0.5$ 

experiment was unclear, but it may be possible that the starting material for this experimental run
included either smaller than average grain sizes or grains with a high degree of internal
fracturing that could have yielded higher surface area to volume ratios for reduction to occur.
Therefore, we conducted more experiments using the same temperature and oxygen fugacity
conditions but with different ilmenite grain sizes to explore the latter possibility.

278 3.4 Reduction experiments with varying grain sizes of starting materials

To study the effect of surface area on the extent of ilmenite reduction and metal creation, we conducted two sets of cooling experiments from 800–500 °C at  $fO_2 = IW-1$  for three different ilmenite grain size ranges (experimental runs H181 and H182). In general, we found that, within each experimental set, the  $M_{rs}$  values decreased with increasing ilmenite grain size for both H181 and H182 (Fig. 4).



284

285

286 Fig. 4: Comparison of the  $M_{\rm rs}$  values of cooling experiments with different grain sizes of starting 287 material. The black squares represent the mean values of  $M_{\rm rs}$  for different grain sizes of starting 288 material from two sets of experiments (run H181 and H182 that were at  $fO_2$ = IW-1 and had a cooling history from 800–500 °C. The grey shadow blocked out the actual  $M_{\rm rs}$  values of small, 289 medium, and large ilmenite grain sizes of experimental runs H181 and H182. 290 291

292 We also compared the other two cooling experiments (experimental runs H171 and H174) 293 and our most reducing isothermal experiment ran at  $fO_2 = IW-2$  (experimental run H158) with 294 experimental runs H181 and H182 (Table 1) to study the interplay between cooling rate and 295 ilmenite grain size effecting on the magnetization properties of the reduction product. 296 Experimental run H174 had the lowest  $M_{\rm rs}$  values; this might be attributable to its high starting

297	temperature at 1000 °C that could have led to the production of larger, more multidomain (MD)
298	metal grains within the sample. Experimental run H171 had the highest $M_{\rm rs}$ values among all
299	experiments. The $M_{rs}$ values for the small (H181_S and H182_S) and medium (H181_M and
300	H182_M) ilmenite grain size experiments were much higher than the experimental run H158's
301	$M_{\rm rs}$ value, and the large ilmenite grain size (H181_L and H182_L) experiments' $M_{\rm rs}$ values were
302	comparable to H158's $M_{\rm rs}$ value (Table 1). Although the oxygen fugacity of H158 was more
303	reducing compared to experimental runs H171, H181, and H182, slow-cooled experiments still
304	showed higher $M_{\rm rs}$ values with smaller grain sizes and comparable $M_{\rm rs}$ values with similar grain
305	sizes. The reduction products, Fe and rutile, were larger and more visible in the BSE images of
306	the slow-cooled experiments too (Fig. 1c and 1d).

307

310

#### 308 4 Discussion

## 309 4.1 Ilmenite subsolidus reduction creates FeNi metal

311 Reduction of ilmenite to rutile and FeNi metals was observed in some Apollo samples. 312 For reduction products in Apollo crystalline rock samples, the metal phases consisted entirely of 313 kamacite (Ahmed El Goresy et al., 1972). There were two steps in the proposed subsolidus 314 reduction reaction: 1) ulvöspinel reduced to ilmenite and metallic iron, and 2) ilmenite reduced 315 to rutile and metallic iron. Intergrowths of ulvöspinel, ilmenite, and metallic iron had been 316 reported in a limited number of Apollo 11 (e.g. Apollo sample 10058-32 described by Cameron, 317 1970) and 12 samples (e.g., Apollo sample 12050 described by Brown et al., 1971; Apollo 318 sample 12020,10 described by Haggerty and Meyer, 1970) and were observed more broadly in 319 Apollo 14 and 17 basalts (e.g., Apollo samples 14053 and 14072 described by El Goresy et al., 320 1971; El Goresy and Ramdohr, 1975; Haggerty, 1971). Ilmenite-ulvöspinel aggregates usually

321 contained native iron, which in most cases was confined to the ilmenite itself or occurred as a 322 compound along the ilmenite-ulvöspinel interface (e.g., Apollo sample 10058-32 described by 323 Cameron, 1970; Apollo sample 15065 described by Taylor et al., 1973). Ilmenite in Apollo 14 324 samples appeared as a primary phase and also through the process of subsolidus reduction of 325 ulvöspinel (Ahmed El Goresy et al., 1972). The Apollo 14 crystalline rocks 14053 and 14072 326 displayed the breakdown of chromian ulvöspinel to a greater extent, with all stages of reduction 327 and complete breakdowns (Ahmed El Goresy et al., 1972). Sample 14072 contained ulvöspinel 328 that was broken down into ilmenite and native Fe. Ilmenite grains and fine native Fe grains had 329 been dissolved along the host's {111} plane (Ahmed El Goresy et al., 1972). Similarly, sample 330 75081 also displayed the thin lamellae of rutile and blebs of metallic iron presenting the ilmenite 331 megacrysts (Taylor, Williams, et al., 1973).

Our experiments were designed to study the effect of temperature, oxygen fugacity, and kinetics of the ilmenite reducing to rutile and metallic iron reduction seen in the Apollo 14 and 17 samples. FeNi metals were produced from our subsolidus reduction experiments, seen in our electron microprobe imaging and quantitative analysis (Fig. 1c, 1d, and Table 3). This was in agreement with the increased  $M_{rs}$  values of the reduced experiments compared to the starting materials (Fig. 3d). In conclusion, both our experimental products and Apollo samples provided evidence for the subsolidus reduction of ilmenite to Fe-metal bearing assemblages.

- 339
- 340

# 4.2 Slow cooling causes ilmenite subsolidus reduction

Apollo mare basalts such as 15495, 15475, and 15065 had coexisting ulvöspinel, ilmenite, and native Fe, and had Zr partitioning indicating the existence of subsolidus reduction of ulvöspinel to ilmenite and native Fe (McCallister & Taylor, 1973; Taylor et al., 1972; Taylor,

McCallister, et al., 1973). It had been proposed that the slow cooling of mare basalts under 900 °C was the reason for the subsolidus reduction from ulvöspinels to ilmenite to rutile and metallic iron (McCallister & Taylor, 1973). The study of the Zr ratio of Apollo samples provided evidence of the relationship between the intensity of reduction and cooling rates (Taylor, McCallister, et al., 1973). For example, 15475 and 15065 showed higher reduction intensity of ulvöspinel than 15495, and the former ones had undergone a slower rate of subsolidus cooling (Taylor, McCallister, et al., 1973).

351 Each of our experiments was run at a constant oxygen offset from the IW buffer curve, 352 but during cooling the absolute  $fO_2$  drops. Slow-cooled experiments produced the highest  $M_{rs}$ 353 values among all of the experiments (e.g., compare experimental runs H171 and H182 to H155 354 and H158) (Table 1 and Section 3.4). Hemingway and Tikoo (2018) also hypothesized that the 355 slow cooling of magmatic features like dikes, sills, and laccoliths could provide the requisite 356 conditions for these subsolidus reduction reaction series to finish. Therefore, in this study, we 357 will testify to the hypothesis of whether these subsolidus reduction reactions in slow cooling 358 conditions can produce enough thermoremanent magnetization for lunar swirls.

359

360 4.3 Slow-cooled hypabyssal dikes provide opportunities for ilmenite subsolidus reduction 361 362 Previous studies had proposed that the formation of lunar swirls was related to dike 363 cooling and the resulting changes in magnetic properties (e.g., Hemingway and Tikoo, 2018). 364 Our experiments confirmed slow cooling rates are the driving force for ilmenite reduction, and 365 we can place constraints on the necessary dike cooling rates for creating magnetizable materials 366 in the lunar subsurface. Our mechanism, subsolidus reduction of ilmenite, may provide an 367 avenue to produce substantial amounts of fine-grained [pseudo-single domain (PSD) or vortex 368 state (Roberts et al., 2017)] metallic iron in lunar rocks. The crustal magnetic field anomalies

369 associated with Reiner Gamma and other lunar swirls could potentially be attributable to the 370 reduction of ilmenite in the cooling of dikes and other intrusive magmatic bodies, but certain 371 conditions need to be met: 1) The appropriately reducing environment must be maintained for a 372 sufficient duration to facilitate the reduction reactions (since this is a diffusion-controlled 373 process), and this could require some slow cooling conditions. 2) The surface area of ilmenite 374 grains needs to be high enough to create enough ferromagnetic materials to account for the 375 observed field strength and estimated magnetization intensity of the crustal magnetic anomalies 376 at swirls. In the following section, we will explain how we used Monte Carlo simulations to help 377 us answer whether our proposed mechanism can produce enough magnetization for lunar swirls.

378 379

4.4 Monte Carlo modeling of the most important three factors affecting the calculated

380 magnetization values

381 Thermoremanent magnetization (TRM) is acquired when rocks cool from above the 382 Curie temperature of their ferromagnetic minerals (e.g., ~770°C for kamacite) in the presence of 383 an ambient magnetic field. We identify three major factors controlling the intensity of TRM that 384 could be acquired as a result of the subsolidus reduction process: 1) the diffusion parameter (f)385 (Section 4.4.1), 2) the domain state of magnetic carriers (Section 4.4.2), and 3) the amount of 386  $TiO_2$  in mare basalts (Section 4.4.2). We note that subsolidus reduction may occur below the 387 kamacite Curie temperature. In such cases, rocks will instead acquire a thermochemical 388 remanent magnetization (TCRM), which may produce a weaker magnetization intensity than a 389 pure TRM. However, because subsolidus reduction largely occurs at high temperatures (>500°C) 390 that are close to the Curie temperature, the difference in the resulting TCRM intensity versus a 391 TRM acquired in the same ambient field strength should be negligible (Draeger et al., 2006). 392 Therefore, for simplicity, we hereafter only discuss TRM.

# 393 4.4.1 Diffusion-controlled ilmenite reduction

394 Our results showed that the ilmenite reduction was driven by a diffusion-controlled 395 reaction progressing from the exterior of the grain to the interior. The reacted zones were 396 concentrated along the surface (grain boundaries and cracks that existed before experiments) (Fig. 397 1b-d). This indicated that the reduction reaction was at least somewhat diffusion controlled. To 398 model this diffusion-controlled process, we used the interdiffusion coefficient of Mg and Fe in 399 ilmenite (Prissel et al., 2020). The experimentally determined Arrhenius fit parameters between Fe and Mg diffusion were 188 kJ mol<sup>-1</sup> for O and -6.0 m<sup>2</sup> s<sup>-1</sup> for  $logD_0$  (Prissel et al., 2020). 400 401 Using the following equation, we calculated the Fe-Mg interdiffusion for our experimental 402 temperature range of 500-800 °C in ilmenite megacrysts.

403

$$\ln D = \ln D_0 - \frac{Q}{RT}$$
 Equation 1

404 where  $D_0$  was the pre-exponential factor (m<sup>2</sup> s<sup>-1</sup>), Q was the activation energy (J mol<sup>-1</sup>), R was 405 the universal gas constant (J mol<sup>-1</sup> K<sup>-1</sup>) and T was temperature (K). The maximum D value was 406  $7.06*10^{-16}$  m<sup>2</sup> s<sup>-1</sup> at 800 °C and the minimum D value was  $1.99*10^{-19}$  m<sup>2</sup> s<sup>-1</sup> at 500 °C.

407

Since ilmenite commonly occurs as a platy crystal in extrusive lunar rocks, we chose to model natural ilmenites by assuming they were infinite plates; approximating that the ilmenite grains were plane sheets. The simple geometry was a good approximation which we verified by also initially modeling crystals as spheres and found the plate geometry was more conservative for reaction progress calculations. The time it would take a platy ilmenite grain with different half-widths to be partially reduced could be approximately calculated by the following equation (McDougall & Harrison, 1999).

415 
$$f \simeq \frac{2}{\sqrt{\pi}} * \sqrt{\frac{D*t}{l^2}}$$
 Equation 2

where *l* was the half-width of the ilmenite (m),  $D (m^2 \text{ s}^{-1})$  was the interdiffusion coefficient between Fe and Mg calculated from Eqn. 1, *f* is the reduction reaction progress, and *t* (s) was the duration for the reduction reaction at different levels of progress. This approximate solution of the diffusion equation applies only when  $f \le 0.60$ . The reduction time of 60% ilmenite with the maximum *D* value was ~0.032 years, and the reduction time of 60% ilmenite with the minimum *D* value was ~113 years.

422

423 The diffusion parameter was affected by the sizes of ilmenites and the widths of dikes. 424 The widths of dikes influenced the necessary cooling time of dikes and thus how much time 425 would be available for the ilmenite reduction reaction to proceed (t). We used the modeled 426 cooling history of a dike of a thickness of 10 m from Snelling (1991) and Jaeger (1957). The 427 dike was intruded as a liquid, and we used these published calculations for a cooling interval of 428 800–500 °C when the dike was below its solidus. The calculated cooling time for a dike of a thickness of 10 m was  $5.37*10^7$  s (~1.73 years). The calculated cooling time for a dike of a 429 430 thickness of 100 m was ~173 years, which was larger than the 60% reduction time with the 431 minimum diffusion coefficient D, ~113 years. The cooling time for a dike of a thickness of 100 432 m from 800–500 °C exceeded 60% of ilmenite reduction time, implying that the cooling time for 433 a dike of a thickness of 100 m was sufficient to reduce ilmenite to rutile and metallic iron.

According to Heiken and Vaniman (1990), the mean  $\pm 1$  standard deviation of the halfwidths of ilmenite grains in high-Ti mare basalts was approximately 53 µm  $\pm 45$  µm. We used these values to represent *l* in Equation 2. More details on how these values were propagated through calculations of TRM would be discussed in sections 4.4.3 and 4.4.4.

438

 $\begin{array}{ll} 440 & 4.4.2 \text{ The domain state of magnetic carriers and the amount of TiO_2 in mare basalts as} \\ 441 & two other important factors \end{array}$ 

442

443 If the subsolidus reduction is a plausible explanation of the subsurface magnetic 444 anomalies associated with the lunar swirls, then the amount of reduction and how much 445 magnetizable material is in slow-cooling dikes, as well as the magnetization recording efficiency 446 of that material are all important factors. In our mechanism, ilmenite reduction is the source of 447 the magnetizable material (metallic Fe or kamacite), and slow-cooling dikes provide the 448 environment for that subsolidus reduction. Thus, the amount of primary TiO<sub>2</sub> in lunar rocks, 449 which dictates how much ilmenite can form, will dictate how much native Fe can be produced. 450 We used the compositions of low-Ti and high-Ti mare basalts to represent two end-member 451 scenarios of Ti contents. The low-Ti basalts tend to have modal mineralogies with  $\leq 2$  vol.% 452 ilmenite (e.g., Papike et al., 1991), whereas there can be up to  $\sim 22$  vol.% ilmenite in high-Ti 453 basalts (Longhi et al., 1974).

454 Magnetizable materials' domain states are also important factors in determining how 455 much TRM they can record. Rock magnetism studies confirmed that Apollo mare basalt samples 456 contain about 0.1 wt.% of predominantly multidomain (MD) FeNi grains (e.g., Fuller, 1974; 457 Strauss et al., 2021). We produced PSD FeNi grains during our slow-cooling subsolidus 458 reduction experiments (Table 3), which might be a better representation of the metal generated 459 within or proximal to slowly-cooled dikes in the lunar crust than more rapidly cooled, surface-460 erupted mare basalts. However, due to the uncertainty in the domain state of true lunar 461 subsolidus reduction products, we used both MD and PSD magnetic carriers to model our results 462 (Section 4.4.3 and 4.4.4).

463

464

4.4.3 How to calculate thermoremanent magnetization  $(M_{tr})$ 

465 To test whether our mechanism can produce sufficient magnetization intensities to 466 explain lunar swirls, we compared the intensities of TRM calculated from different variables: 467 domain states of magnetic carriers, TiO<sub>2</sub> contents in lunar rocks, and lunar dike cooling rates. 468 We followed the methods of Oliveira et al. (2017) to calculate our experiments' and mare basalts' TRM. Briefly,  $M_{tr}$  can be calculated from Equation 3 if we know the following variables: the 469 470 volumetric concentration of the magnetic carrier c, the saturation magnetization of metallic iron  $M_{\rm s}^{\rm Fe}$ , the squareness ratio s of the hysteresis loop, the applied magnetic field B, and the constant 471 472 а.

473 
$$M_{\rm tr} = \frac{c \, s \, B \, M_{\rm s}^{\rm Fe}}{a}$$
 Equation 3.

where  $M_s^{\text{Fe}}$  was 1.715 \* 10<sup>6</sup> A m<sup>-1</sup> (Dunlop & Özdemir, 2015), and c was the volumetric 474 475 concentration of Fe reduced from low-Ti and high-Ti mare basalts. The amounts of metallic Fe 476 can be estimated by the reduction of ilmenite from low-Ti and high-Ti basalts using mass 477 balance and densities. B ranged from 10  $\mu$ T to 100  $\mu$ T because ~100  $\mu$ T was the upper limit of 478 lunar paleointensities that had been inferred from paleomagnetic studies of Apollo samples. 479 Constant a varied from ~2810  $\mu$ T for multidomain samples to ~3770  $\mu$ T for single-domain and 480 pseudo-domain samples (Weiss & Tikoo, 2014; Wieczorek et al., 2022). The squareness s was the ratio of the saturation remanent magnetization  $(M_{rs})$  and saturation magnetization  $(M_s)$  of the 481 482 experimental samples and mare basalts. To understand the boundaries between domain states, 483 we assumed  $M_{rs}/M_s < 0.05$  was MD; between 0.05 and 0.5 was PSD, and >0.5 was SD (see Fig. 484 7 of Strauss et al. 2021).  $M_{rs}/M_s$  values from our experiments are listed in Table 1. We selected 485 the experiments that were conducted under  $fO_2 = IW-1$  and IW-2 and there were EPM analyses on the metallic irons (Table 3). The mean  $M_{\rm rs}/M_{\rm s}$  value from these experiments was 0.295 486 487 (PSD). We found that FeNi grains formed during our subsolidus reduction experiments were on

488 average smaller (pseudo-single domain;  $M_{rs}/M_s \sim 0.1$ ) (Table 1) than those naturally occurring within mare basalts (multidomain;  $M_{\rm rs}/M_{\rm s}$  ~0.001-0.01). A typical  $M_{\rm rs}/M_{\rm s}$  value for mare 489 490 basalts was ~0.0064 (Fuller & Cisowski, 1987). This was of interest because pseudo-single 491 domain grains can more efficiently record thermal remanent magnetization than multidomain grains for a given ambient field intensity (Fig. 5). We noted that in a natural setting, protracted 492 493 cooling on timescales far exceeding the durations of our laboratory experiments may result in the 494 growth of MD grains rather than PSD grains. While it would be desirable to conduct an 495 experiment analogous to what could occur on the Moon, such long timescales (~100+ years to 496 cool a 100 m dike) is beyond the capability of our analysis.



498 Fig. 5: Different scenarios of percentages of calculated  $M_{tr}$  values that are over the minimum 499 requirement for lunar swirls (0.5 A/m) with magnetizing field ranging from 10–100  $\mu$ T with a 500 dike width = 100 m. Blue line: high-Ti basalts with pseudo-single domain magnetic carriers;

- 501 blue dashed line: high-Ti basalts with multidomain magnetic carriers; black line: low-Ti basalts
- 502 with pseudo-single domain magnetic carriers; black dashed line: low-Ti basalts with
- 503 multidomain magnetic carriers.

504 4.4.4 Variables were artificially generated to explore a significant proportion of the 505 realistic parameter space

506 As discussed in the previous section, there are many variables that we do not have precise 507 constraints on because 1) they vary in nature and/or 2) there are uncertainties in the lab analysis. 508 In order to handle uncertainties and variabilities, our strategy was to generate artificial datasets 509 utilizing MATLAB and Monte Carlo simulations to explore the full range of possible parameters. We used lognormal distributions and Monte Carlo simulations to generate  $10^6$  random 510 511 numbers to represent the half-widths of ilmenites (1) in Equation 2 based on the average and one 512 std values of the half-widths of ilmenite grains in high-Ti mare basalts. We used Monte Carlo simulations to find 10<sup>6</sup> uniformly distributed random numbers for each of the following variables: 513 514 (1) calculated reaction completeness parameter (f) from Equation 2; (2) applied magnetic field B 515 from 10  $\mu$ T to 100  $\mu$ T; and (3) two sets of numbers to represent volume percentages of ilmenites 516 one each for low-Ti and high-Ti mare basalts. The ilmenite volume percentages were 0.5-5 vol.% 517 for low-Ti mare basalts and 9-25 vol.% for high-Ti mare basalts. With our current knowledge 518 on the chemical and physical properties of lunar rocks and dike cooling histories, we believed 519 that utilizing Monte Carlo simulation provides an optimal approach to handling variables when 520 calculating the predicted TRM intensities of lunar rocks.

We assumed that <60% of ilmenite was reduced to rutile and metallic Fe (Section 4.4.1) because not all ilmenite had been reduced to rutile and Fe in Apollo samples (Taylor, Williams, et al., 1973). Therefore, we only used *f* values between 0 and 0.6 calculated from Equation 2 to simulate how much ilmenite in either low-Ti or high-Ti mare basalts was reduced to rutile and Fe metals. Another assumption was that the ilmenite here was only FeTiO<sub>3</sub>, but there might be a 526 small amount of MgO existing in the ilmenite (e.g., Alexander et al., 2016; Papike et al., 1991).

527 These assumptions allow us to explore a significant proportion of the realistic parameter space.

528

529 4.5 Our proposed reduction mechanism can reproduce lunar swirls' surface magnetic530 anomalies

As we modeled the effects of the reaction completeness parameter (f), the domain state of magnetic carriers, and the amount of TiO<sub>2</sub> in mare basalts on the reduction of ilmenites in the previous section, now we can answer the question of whether this diffusion-controlled subsolidus reaction can produce enough magnetization for lunar swirls.

535 The intensity of TRM will increase with both higher metallic Fe content as well as with 536 finer average grain sizes (Fig. 5). Lunar swirls were inferred to need magnetization intensities 537 of >0.5 A/m, which could easily be achieved for higher Ti basaltic rocks magnetized in fields of 538 a few microteslas or stronger. According to Hemingway and Garrick-Bethell (2012), the 539 remanent crustal field observed at Reiner Gamma requires an associated source magnetization 540 intensity of 1–100 A/m, depending on source geometry, whereas Airy and other swirls could be 541 explained by somewhat weaker magnetization intensities. Our Monte Carlo simulations reveal 542 several different scenarios that can produce TRM intensities exceeding the minimum 0.5 A/m 543 threshold value (Fig. 5).

For dikes with a low-Ti mare basalt composition and MD FeNi grains, around 20% of calculated TRM reached over 0.5 A/m at around 40  $\mu$ T ambient fields and if the half-width of ilmenite was within favorable ranges (Fig. 5). However, we noted this was only the case for a small fraction of model runs. In contrast, for low-Ti dikes with PSD FeNi grains, around 80% of

our Monte Carlo simulations could reach over 0.5 A/m TRM intensities at around 20 μT ambient
field (when varying both the dike cooling time and half-widths of ilmenite) (Fig. 5).

There were more possible pathways for the dikes with high-Ti mare basalt compositions to meet the minimum magnetization threshold to explain lunar swirls. For this lithology, ~18% of Monte Carlo runs using rocks with MD grains could produce TRM >0.5 A/m if the paleofield was >10  $\mu$ T (when varying the half-widths of ilmenite) (Fig. 5). For Monte Carlo runs employing high-Ti mare basalt compositions and PSD FeNi grains, almost all the resulting TRM intensities exceeded 0.5 A/m (even for paleofields as low as 10  $\mu$ T) when both the dike cooling rate and half-width of ilmenite were varied (Fig. 5).

557 We note that our minimum 0.5 A/m magnetization intensity is much lower than some 558 estimates for the magnetization intensity at the Reiner Gamma swirl (e.g., >10 A/m from 559 Garrick-Bethell and Kelley, 2019). We note that based on the magnetic properties of known 560 lunar rocks, it is not possible to obtain TRM intensities >10 A/m, even for lunar impact melt 561 breccias or melt rocks, from a dynamo field with <100 microtesla intensity (D. J. Hemingway & 562 Tikoo, 2018). The incorporation of iron derived from metal-rich impactors is the most likely 563 scenario to produce >10 A/m magnetization intensities in the lunar crust (D. J. Hemingway & 564 Tikoo, 2018). Therefore, our work narrows down the origin of lunar swirl magnetic source 565 bodies to either impactor ejecta or our hypothesis of subsolidus reduction of dikes/sills with the 566 caveat that this reduction must occur under certain circumstances that may not be exceedingly 567 common (most favorably, a combination of high-Ti initial compositions, PSD domain states, and 568 strong paleofield intensities). Testing between the subsolidus reduction of magmatic intrusive 569 versus impactor metal hypotheses will require making high-resolution lunar crustal magnetic 570 field measurements during surface or near-surface traverses to better constrain the geometries of

571	the magnetic source bodies. The upcoming Lunar Vertex rover, which will visit Reiner Gamma
572	in 2024, presents the nearest-term opportunity to conduct such an investigation (D. T. Blewett et
573	al., 2022).

574

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515	3	COL	usions	5

- 576
- 577 1. The slow cooling and subsolidus reduction of lunar magmatic intrusive bodies is a578 plausible mechanism for producing intense lunar crustal magnetism.
- 579 2. The ilmenite reduction reaction is a diffusion-controlled reaction; thus, it is dependent on 580 temperature, initial ilmenite grain sizes, and  $fO_2$ .
- 3. Specifically, reduction within hypabyssal dikes with high-Ti or low-Ti mare basalt
  compositions can produce sufficient FeNi grains to carry the minimum >0.5 A/m
  magnetization intensity inferred for swirls, especially if ambient fields are >10 μT or if
  fine-grained Fe-Ni metals in the pseudo-single domain grain size range are formed (Fig.
  5). Due to their higher ilmenite content, reduction of high-Ti mare basalts can more
  easily produce >0.5 A/m magnetizations than reduction of low-Ti mare basalts.
- 587
  4. Our study highlights that the domain state of the magnetic carriers is an under588 appreciated factor in controlling a rock's magnetization intensity. This can be even more
  589 important than the metal content of lunar rocks.
- 590 5. It is indeed possible for the lunar swirl magnetic anomalies to form from an endogenic 591 origin; however, it may require a specialized combination of initial conditions (i.e., dike 592 width, initial ilmenite content, magnetic domain state, ambient field intensity) that may 593 not commonly occur in tandem.
- 594 6. Upcoming missions that will traverse lunar swirls while conducting magnetic field
   595 measurements will further elucidate the origin of swirl magnetic source bodies.

596

# 597 **Open Research:**

# 598 **Data Availability Statement**

We have shared our raw EPMA and Rock Magnetic data at the following link: https://doi.org/10.7936/6RXS-103643. In the WashU Research Data repository, there are .cvs files of RawEPMAData that contain raw EPMA data for the experiments published in the manuscript. RawRockmagData.cvs files contain raw magnetic hysteresis and raw demagnetization data for the experiments published in the manuscript. The raw EPMA and rock magnetization data meet the principles of FAIR data (findable, accessible, interoperable, & reusable).

606

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