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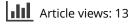
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# Characterisation of carbonate minerals from hyperspectral TIR scanning using features at 14 000 and 11 300 nm

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#### ABSTRACT

Rapid characterisation of carbonate phases in hyperspectral reflectance spectra acquired from drill core material has important implications for mineral exploration and resource modelling. Major infrared active features of carbonates lie in the thermal region around 6500 nm, 11 300 nm and 14 000 nm, with the latter two features being most useful for differentiating mineral species. A scatter diagram of the wavelength of the 14 000 nm feature *vs* that of the 11 300 nm feature, powerfully differentiates carbonates. Although the wavelength of the 11 300 nm peak is easily measured, the 14 000 nm trough and peak are commonly weak and their wavelengths can confidently be used only after filtering the spectra, e.g. selecting only those with the trough and peak separated by 175–230 nm, typical of common carbonates. The method is demonstrated with drillhole 120R from the Rosebery polymetallic VHMS deposit in western Tasmania, which has been scanned with the HyLogger-3 system. A 14 000–11 300 plot shows a high degree of clustering of the drillhole 120R data close to the library spectra of calcite, dolomite, Fe-dolomite, ankerite, kutnohorite, rhodochrosite, Fe-rhodochrosite and siderite. The interpreted compositions of the carbonate spectral populations strongly correlate with the chemical populations of 144 analysed carbonates and provide a highly resolved spatial framework for interpreting carbonate alteration.

# Introduction

Differentiation of carbonate minerals using hyperspectral infrared reflectance spectroscopy is limited by the wavelength range, intensity and shape of their spectral features (i.e. vibrational modes) and the ability to distinguish these from overlapping features of other minerals. The interpretation of visible–near-infrared (VNIR, 350 to 1000 nm) and short wave infrared (SWIR, 1000 to 2500 nm) reflectance spectra is limited by the availability of only one major spectral feature for most carbonates. This problem is addressed when using thermal infrared (TIR) spectra, for example from the HyLogger-3<sup>TM</sup> core scanning system (6000 to 14 500 nm), which typically contain three carbonate spectral features, potentially offering an improved capability for interpretation.

This paper briefly reviews the nature and characteristics of the spectral features of carbonates and introduces a practical method for using the wavelengths of diagnostic, but noisy TIR features at 14 000 nm together with a feature at 11 300 nm to differentiate carbonate minerals. The method typically requires heavy filtering of spectra and is therefore most applicable to voluminous hyperspectral data such as that of the National Virtual Core Library. The method is applied to a case study of HyLogger-3<sup>TM</sup> data from the Rosebery base metal deposit in western Tasmania.

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Thermal infrared; carbonate; HyLogger; spectral geology; Rosebery; volcanic-hosted massive sulfide

#### Rosebery drillhole 120R

TIR hyperspectral scanning has great potential to advance the understanding of alteration systems in which carbonates play an important role. For example, the world-class Rosebery polymetallic volcanic-hosted massive sulfide (VHMS) deposit in western Tasmania comprises 19 stratabound base metal ore lenses surrounded by zoned alteration envelopes comprising chlorite, sericite and carbonates in a guartz-albite-phengite-chlorite, chiefly volcaniclastic host sequence (McNeill, 2014). Although it is generally recognised that the Mn-carbonates rhodochrosite MnCO<sub>3</sub> and kutnohorite (CaMn)CO3 are essential components of diagenetic and ore-related alteration and that calcite occurs in later veins (Allen, 1997), the compositional modes and spatial distributions of carbonate species are not well known. Drillhole 120R provides a well-studied (Herrmann et al., 2001; Large, Allen, Blake, & Herrmann, 2001) complete section through the Rosebery mine stratigraphy and alteration and has been scanned by the HyLogger-3 system, providing suitable high spatial and spectral resolution data for detailed carbonate mapping.

# Carbonate VNIR and SWIR features

In the SWIR region, carbonates are typically characterised using the wavelength of the 2340 nm absorption feature (e.g. Gaffey,

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1986a), ascribed to the 2nd overtone of the  $\nu$ 3 asymmetric stretch of the CO<sub>3</sub> anion (Workman & Weyer, 2008). Although the feature has been found to be useful in differentiating magnesite, dolomite and calcite (Gaffey, 1986a; Zaini, van der Meer, & van der Werff, 2012), it is less useful for distinguishing Febearing carbonates from those dominated by Ca, Mg and Mn (AusSpec International, 2008; Clark, 1999; Gaffey, 1986b).

A discriminator for Fe-bearing carbonates is the strength (and structure) of the Fe<sup>2+</sup> crystal field absorption bands in the near-infrared region at  $\sim$ 1200 nm, attributed to electronic transitions of the Fe<sup>2+</sup> ion (Gaffey, 1986b). In practice, the strength of the Fe<sup>2+</sup> crystal field feature has been found to be highly variable, especially when comparing across mineral species (Gaffey, 1986a; Hunt & Salisbury, 1971), reducing its value in discriminating sideritic carbonates.

#### **Carbonate TIR features**

Diagnostic spectral features of carbonates in the TIR region (Figure 1; Table 1) are caused by fundamental vibrations of the CO<sub>3</sub> anion and can be separated into three different types (Lane & Christensen, 1997):  $\nu$ 2 out-of-plane bend,  $\nu$ 3 asymmetric stretch and  $\nu$ 4 in-plane bend.

The dominantly  $\nu$ 3 peak at 6500 nm has a complicated shape owing to superposition of  $\nu$ 3 with the harmonics of  $\nu$ 2 and  $\nu$ 4 and therefore has the potential to contain many diagnostic characteristics. However, only the wavelength of the major peak

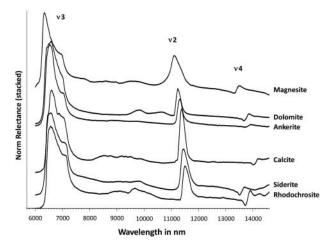


Figure 1. Stacked carbonate TIR spectra illustrating the diagnostic spectral features.

	v3 peak	v2 peak	v4 trough	v4 peak
Magnesite MgCO <sub>3</sub>	6405	11058	13235	13433
Dolomite (Ca, Mg)CO <sub>3</sub>	6490	11236	13605	13829
Ankerite (Ca, Fe>Mg)CO <sub>3</sub>	6497	11290	13656	13837
Calcite CaCO <sub>3</sub>	6598	11343	13942	14129
Siderite FeCO <sub>3</sub>	6566	11427	13417	13614
Rhodochrosite MnCO <sub>3</sub>	6549	11483	13638	13837

Accuracy of feature positions for calcite: v2 (11 343 nm) =  $\pm$  25 nm; v3 (6598 nm) =  $\pm$  73 nm; v4 (14 129 nm) =  $\pm$  15 nm.

of the group is commonly used in discriminating between carbonates (e.g. Huntington, 2006; Huntington et al., 2011). The feature has a wavelength range of 220 nm and has large uncertainty owing to the broad (>1000 nm) and commonly undulating shape of the peak crest. Chester and Elderfield (1966) suggested that the  $\nu$ 3 peak is not suitable for distinguishing individual carbonate minerals, as it is located at approximately the same wavelength position in several carbonate phases. The 6500 nm feature is also particularly subject to significant modification by volume scattering when the grainsize is small (Lane, 1999; Myers et al., 2015; Zaini et al., 2012).

The most diagnostic carbonate features in TIR reflectance spectra are the  $\nu$ 2 peak at 11 300 nm, which ranges over 360 nm, and the  $\nu$ 4 peak at 13 200–14 100 nm and adjacent trough, which each range over 730 nm. At hyperspectral resolution, the 11 300 nm peak is typically prominent in the TIR and is sharp, with an FWHM of less than 400 nm. The 14 000 nm features span the greatest wavelength range of all carbonate infrared spectral features and are narrow, each with an FWHM less than 300 nm, and are therefore potentially the most discriminating. However, the 14 000 nm features are commonly weak and difficult to confidently identify, especially when there are additional minerals in the assemblage that have features in the same region.

The trough and adjacent peak at 14 000 nm, characteristic of carbonates, are separated by 160–280 nm, with most common carbonates within the range 175–230 nm (Figure 2). This

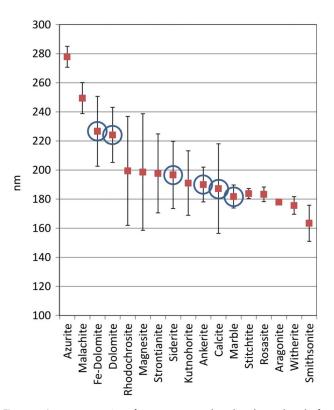


Figure 2. Average separation of 14 000 nm trough and peak wavelengths for carbonates in the TSG spectral library (Schodlok et al., 2012, 2016b) plus additional ankerite and kutnohorite spectra from western Tasmanian drillhole samples as described in the text. The most common carbonates are circled, and the vertical bars indicate the 1 standard deviation uncertainty limits.

# Methods

The spectral reference library of Schodlok, Warren, Green, and Huntington (2012) and Schodlok, Green, and Huntington (2016b), developed for the HyLogger drill core logging system, was used to characterise carbonate spectral features. The reference samples are pure, solid, chiefly museum grade minerals and bi-directional reflection spectra were collected using a HyLogger-3 system. A virtually identical HyLogger-3 system at Mineral Resources Tasmania, Hobart, Tasmania, which gathers data for the National Virtual Core Library, was used to augment the reference library with additional ankerite and kutnohorite spectra from western Tasmanian drillhole samples (drillholes MJ021, 648.15m and drillhole 120R, 1365.88m, validated by XRD at Mineral Resources Tasmania, Hobart). This HyLogger was also used to scan the case study drillhole. In scanning mode, the instrument acquired co-registered visible, SWIR and TIR spectra, with a spectral resolution of 25 nm, from slightly overlapping samples spaced 8 mm along drill core (Schodlok et al., 2016a). Automatic processing of the raw data by The Spectral Geologist<sup>™</sup> (TSG) software produced corrected reflectance spectra (Schodlok et al., 2016a).

The spectral indices of interest (i.e. peak and trough height and wavelength of carbonate features) of library and case study drill core samples were measured using the scalar index tools in TSG (detailed in Table 2). The presence of carbonate in spectra was signalled by a peak at 11 300 nm, with >0.01reflectance relative to the hull envelope. In order to then select only those carbonate-bearing spectra (either pure or within a mineral mixture) with the most reliable 14 000 nm features, the trough and peak wavelengths and their feature separation were calculated, and only those with a feature separation between 175 and 230 nm were retained. Only after this critical step can the actual wavelength of a 14 000 nm feature be used (together with the wavelength of the 11 300 nm feature) to discriminate between carbonate minerals. Although either the 14 000 nm peak or adjacent trough wavelength could be equivalently be used as the diagnostic 14 000 nm feature, for HyLogger-3 data the wavelength of the 14 000 nm trough was preferred, as the adjacent peak is located a little nearer the wavelength limit of the system and may be subject to more noise. Albite has a peak and adjacent trough near 14 000 nm separated by 180 nm, similar to calcite, so carbonate-bearing spectra with significant albite (>0.2 spectral weight proportion, determined using constrained least-squares unmixing; Schodlok et al., 2016a) were excluded from further analysis.

The Rosebery deposit is characterised by carbonate alteration including many of the carbonate minerals included in the reference library: calcite, dolomite, ankerite, siderite and rhodochrosite. Drillhole 120R, which intersects the carbonate alteration, has been scanned by the HyLogger-3 system and processed to optimise imagery, mask gaps in the core, unmix spectra to mineral end-members using the constrained leastsquares method (Schodlok et al., 2016a), and calculate the required spectral indices (detailed in Table 2). The drillhole provided 411 m, or 48 000 rock spectra, including 4635 carbonate spectra, to compare with those of the library carbonates. Historic microprobe analyses of 144 carbonates from the drillhole (Large, Allen, & Blake, 1997) provided data for validating spectral interpretations of carbonate mineralogy.

## Results

#### Library carbonates

The two most discriminating carbonate spectral features, the 14 000 nm trough wavelength and the 11 300 nm peak wavelength, are used to create an optimally diagnostic diagram (14 000–11 300; Figure 3). In this plot the library minerals are well scattered, with the Ca, Mg, Fe and Mn end-members significantly separated from each other and the dolomite-ankerite series defining a linear array. The rhodochrosite spectra appear to define an end-member rhodochrosite and an Fe-rhodochrosite.

#### Rosebery drillhole 120R

In the 14 000–11 300 diagnostic diagram, populations of HyLogged carbonate spectra from drillhole 120R cluster around the positions of the calcite, rhodochrosite and

Table 2. Practical scalars to filter and measure carbonate 14 000 nm features from reflection spectra. The nomenclature relates directly to The Spectral Geologist software, but the methods can be applied using other software. Albite is identified using the constrained least-squares unmixing method (TIR-CLS, Schodlok et al., 2016b), but other unmixing algorithms such as TSG's The Spectral Assistant<sup>™</sup> (Mason & Bischof, 1997) or tailored spectral indices could be used. The final filter scalar (a mask) uses a combination of several simple Boolean scalars derived from scalars 1 to 5. The scalars that include filtering criteria are 2, 3 and 6.

Scalar	Method	Target	Height $\leq$	Local continuum	Fitting interval	Focus interval	Polynomial order	Result
Cb 11 300 height	PFit	Peaks, maxima	0	Hull envelope, subtract	10 900-11 800	11 000–11 650	10	Relative height
Cb 14 000 trough wvl	PFit	Troughs, minima	0.005	Hull envelope, subtract	13 200-14 200	13 380-14 100	12	Wavelength
Cb 14 000 peak wvl	PFit	Peaks, maxima	0.005	Hull envelope, subtract	13 250-14 300	13 400-14 200	12	Wavelength
Cb 14 000 peak-trough	Arith	Cb 14 000 peak w	vl–Cb 14 00	0 trough wvl				5
Ab CLS wt	ClassEx	Search TIR-CLS Mi	inerals for A	lbite, extracting correspon	nding TIR-CLS we	ight (changing N	IULL match re	sult to zero)
Cb with useful 14 000	UClass	Create mask from			5	5 . 5 5		
		(Cb 11 300 height ≤230)	: > 0.01 AN	D Ab CLS wt $<$ 0.2) AND (	Cb 14 000 peak-t	rough $\geq$ 175 AND	0 Cb 14 000 pe	eak-trough

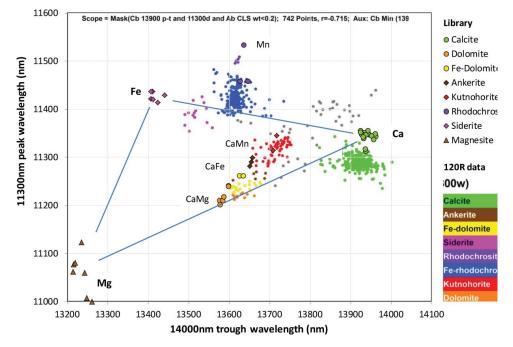


Figure 3. Carbonate discrimination scatter plot of common library carbonate minerals (larger labelled symbols, with Ca, Mg and Fe end-members connected by blue lines). The data for drillhole 120R carbonate-bearing spectra are also plotted (small circles), coloured by proximity to the labelled library minerals, with ambiguous data represented by the grey NULL category.

Fe-rhodochrosite reference minerals (Figure 3). Another linear array with varying Mg, Fe and Mn corresponds to the dolomite to ankerite series. A small population distinct from Ferhodochrosite plots close to siderite.

Electron microprobe analyses of drillhole 120R carbonates (from Large et al., 1997) are located to within 10–100 cm, so cannot be matched with individual HyLogger-3 spectra, but agree with the spectral interpretation with three groupings: calcite, bimodal rhodochrosite and Fe-rhodochrosite, and a continuous series from ankerite to kutnohorite (Figure 4). The microprobe data include one siderite at 1200 m depth,

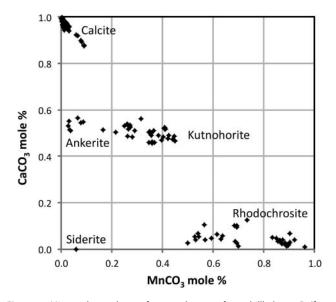


Figure 4. Microprobe analyses of 144 carbonates from drillhole 120R (from Large et al., 1997).

corresponding to a small population interpreted from the spectral data.

# Discussion

## **Diagnostic diagrams**

The results clearly demonstrate the diagnostic power of the 14 000–11 300 scatter diagram. Library spectra are well separated with topological relationships generally honouring chemistry, e.g. the dolomite–ankerite series appears as a linear array between calcite and the siderite to magnesite tie line. Although rhodochrosite lies outside the Ca–Mg–Fe composition space, kutnohorite plots on the ankerite side of a tie line between calcite and rhodochrosite, indicating either a non-linear spectral response or the presence of Mg. The ankerite analysed from drillhole 120R has an Fe:Mg mole proportion averaging 66:33, consistent with its location relative to end-members in Figure 3 and the spectral composition of Ca–Mg–Fe carbonates being linearly related to the chemical composition, as suggested by Chester and Elderfield (1966).

The relative benefit of using the 14 000–11 300 plot is illustrated when the library minerals are plotted on alternative diagnostic diagrams, e.g. TIR 11 300–6500 and SWIR 2340–  $Fe^{2+}$  intensity (Figure 5). In the TIR plot, the library spectra for each mineral species are poorly clustered, rendering classification ambiguous. The mineral species also tend to lie along a line, indicating that the two spectral features are correlated, weakening their combined discriminatory power. In the SWIR plot, the library spectra are largely sensitive only to the 2340 nm feature, with Fe-bearing carbonates plotting inconsistently, as found by Gaffey (1986a), who showed that a deep

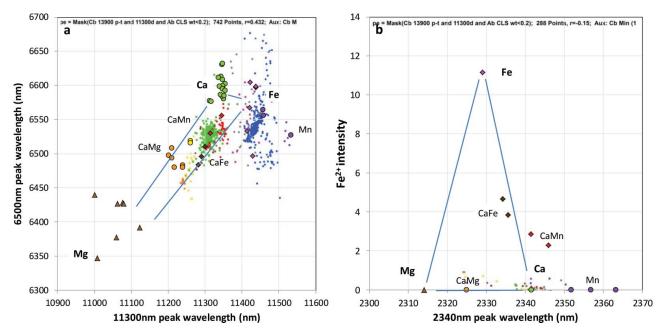


Figure 5. Alternative carbonate discrimination scatter plots showing library minerals and data from drillhole 120R, both symbolised as in Figure 3: (a) TIR 11 300– 6500 and (b) SWIR 2340– $Fe^{2+}$  intensity (the latter calculated as the sum of the reflectance gradients on either side of the 1200 nm absorption feature, at 760– 1150 nm and 1300–1760 nm). The TIR data have been filtered using height of the 11 300 nm peak to provide the same number of spectra as in Figure 3.

 $Fe^{2+}$  absorption feature can be produced by a dolomite with only 3.26 wt% Fe (dolomite<sub>95</sub>siderite<sub>5</sub>).

In the alternative diagnostic diagrams (Figure 5), the data from drillhole 120R are clustered into only one or two populations and these are difficult to associate with library spectra. In the SWIR 2340–Fe<sup>2+</sup> intensity plot, the dolomite– ankerite–kutnohorite population is not separated from calcite. In the TIR 11 300–6500 plot, the 6500 nm feature of calcites consistently plots with an anomalously low wavelength, characteristic of volume scattering, to which this feature is particularly susceptible (Bishop et al., 2013; Lane, 1999; Lane & Christensen 1998). In summary, the alternative diagnostic plots are unsatisfactory owing to (i) poor correspondence to library spectra and (ii) relatively weak clustering of the drillhole data, at least in part owing to inaccurate or compromised determination of relatively poorly characterised spectral variables.

#### Spatial variation

Plotting the carbonate mineralogy interpreted from Figure 3 down the drill hole shows that siderite is localised at the ore zone and in veins within black shale near 1200 m depth (Figure 6a, b). An Fe-rhodochrosite zone at and above the ore, but bounded above by quartz rhyolite, contains rhodochrosite at its core and is fringed by kutnohorite. Dolomite is restricted to black shale units, and Fe-dolomite and ankerite are progressively more widespread. Calcite occurs throughout the hole but is rare in the footwall pumice breccia. The spatial interpretation is consistent with the downhole distribution of analysed compositions (Figure 6c) and provides much improved data continuity and spatial resolution relative to existing geological logging and microprobe data.

#### Volume scattering

Carbonate spectra exhibiting strong volume scattering effects are characterised by 6500 nm and 11 300 nm features modified by nearly superimposed absorption troughs, resulting in weaker reflection peaks, with maxima displaced from their unmodified wavelengths (e.g. Lane & Christensen, 1998; Roush, 2010). The 14 000 nm features are less susceptible to similar modification (Lane, 1999). It is evident in Figures 3 and 5a that most drillhole 120R calcites plot with 14 000 nm features corresponding to library spectra, but with 11 300 nm and 6500 nm features plotting 35 nm and 50 nm less than the average library spectra. A correlation between 6500 nm peak height and wavelength (both decreasing) is consistent with the calcite spectra being modified by volume scattering.

A small population of drillhole 120R spectra plot with 14 000 nm features corresponding to calcite, but with an anomalously long 11 300 nm peak wavelength (grey symbols in Figure 3). These spectra suffer from strong volume scattering effects, with greatly weakened 6500 nm peaks. A few spectra plotting close to kutnohorite, but with anomalously long 11 300 nm peak wavelengths, also show similar volume strong scattering effects. A common cause of strong volume scattering is fine grainsize (Lane & Christensen, 1998; Roush, 2010), but both the calcite and kutnohorite showing strong scattering effects do not appear to be anomalously fine-grained in the drill core. Another possible cause that can not be discounted is surface coating by fines.

#### Testing under-representation

The process of filtering carbonate spectra to select only those with measurable 14 000 nm features has the potential to

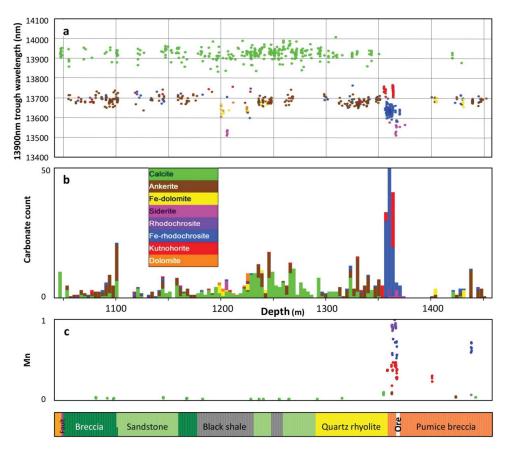


Figure 6. Data from drillhole 120R: (a) Downhole scatter graph of the diagnostic wavelength of the carbonate 14 000 nm trough feature coloured by the mineral interpretation as in Figure 3. (b) Downhole histogram of carbonates coloured by the spectral mineral interpretation. The y-axis has been clipped from a maximum of 103. (c) Mn content (mole fraction) of analysed carbonates, coloured by chemical mineral identification, showing spatial consistency with the spectral interpretation. The lithological log is simplified from Allen, Duhig, and Large (1996).

leave some carbonate populations under-represented in the classification. These can be sought by relaxing the filtering criteria (Table 2) and examining any arising concentrations of data in the diagnostic 14 000–11 300 scatter plot and in downhole distributions. In drillhole 120R, although only 20% of carbonate-bearing spectra survived filtering, no additional populations were found when the filtering criteria were progressively relaxed.

# Conclusions

The most diagnostic visible, SWIR or TIR hyperspectral features of carbonate minerals are the wavelengths of the 14 000 nm and 11 300 nm features. A scatter plot of these two spectral indices separates library mineral spectra widely, with intermediate Ca–Fe–Mg compositions indicating linear spectral mixing. Data from the Rosebery 120R drillhole plot close to library minerals, enabling confident mineral identification.

Although the 14 000 nm trough and peak are the most powerfully diagnostic of all carbonate spectral features, they are typically weak and susceptible to noise and interference from other minerals. When interpreting natural data such as that from drillholes, spectra should be filtered to extract only those spectra with the most reliable 14 000 nm features. A useful filter selects spectra with an adjacent trough and peak at 14 000 nm separated by 175–230 nm, a characteristic feature of most carbonates. Although only a proportion of carbonate spectra contain 14 000 nm features that can be confidently identified and measured, the large number of spectra produced by hyperspectral scanning systems such as the HyLogger-3 nevertheless provide sufficient spatial and compositional completeness. Analysis and validation of carbonate spectra from HyLogger-3 scanning of drillhole 120R from the Rosebery VHMS deposit demonstrate the discriminatory power of TIR spectral analysis and illustrate the statistical robustness and remarkable spatial detail resulting from spectral scanning.

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# **Disclosure statement**

No potential conflict of interest was reported by the authors.

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