2026 International Nuclear Atlantic Conference – INAC 2026
Escola de Guerra Naval, Rio de Janeiro, RJ, August 24 - 28, 2026

ISBN: 978-65-272-2398-6

Investigating the origin of lapis lazuli samples for applications in high-dose dosimetry

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1. Introduction

Lapis lazuli is a metamorphic rock of high commercial and historical value, used since antiquity in ornaments and pigments due to its intense blue coloration. Parallel to its market appeal, the study of complex silicates has aroused great interest in radiation physics for its potential application in high-dose dosimetry, since the presence of crystalline defects and impurities in these minerals acts as charge-trapping centers fundamental to luminescent processes [1, 2]. However, the precise identification of the geographical origin of lapis lazuli samples represents a complex challenge in mineralogy, given that the secondary constituents vary subtly depending on the formation environment. Faced with this difficulty, this study proposes to investigate the material from two distinct sources declared by the sellers: Afghanistan and Mexico, through analytical characterization techniques such as: X-ray Fluorescence (XRF), X-ray Diffraction (XRD), and Electron Paramagnetic Resonance (EPR), aiming to map their compositional and structural signatures.

2. Methodology

The samples were crushed and sieved into grain sizes of 75 μm, 63 μm and < 45 μm. The chemical composition was determined by XRF, using samples with a grain size of 63 μm on a benchtop spectrometer by the fundamental parameters method (7 kV, 60 s, He atmosphere) [3]. Crystalline phases were identified by XRD using Cu-Kα radiation between 5° and 90° (2θ) for phase identification via the PDF-ICDD database and the application of Bragg's Law ($n\lambda = 2d \sin\theta$) [4]. EPR spectra were obtained using a Bruker EMXplus spectrometer operating in the X-band for the calculation of the g-factor ($g = \frac{h\nu}{\mu_B\ Br}$) at room temperature, allowing the evaluation of signal intensity, linewidth, and paramagnetic behavior [5].

3. Results and Discussions

The integration of analytical techniques revealed markedly distinct geological signatures, allowing the correlation of mineralogy with the origin of the samples.

Elemental Signature - XRF: The sample from Afghanistan revealed an essentially silicate nature, with a high content of SiO_2 (52.30%) and Al_2O_3 (11.35%), suggesting an abundance of quartz and aluminosilicates. On the other hand, the sample from Mexico presented a predominantly carbonate character, with a significant concentration of CaO (40.98%) and MgO (8.79%), indicating the majority presence of calcite and dolomite.

Crystalline Phases - XRD: The diffractograms confirmed the chemical inferences (Figure 1). The Afghan sample exhibited intense peaks of silicate phases, while the Mexican material was identified as diopside (monoclinic pyroxene space group, *C12/c1*) [6], consistent with partial substitution of Mg^{2+} by Ni^{2+} within

the crystal lattice.

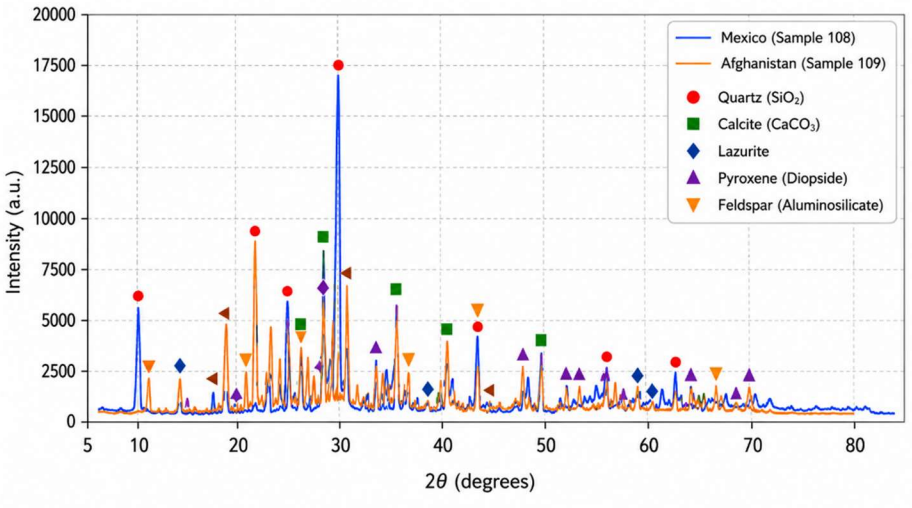


Figure 1 – Correlation of XRD Diffractograms of samples from Afghanistan and Mexico

Paramagnetic Centers - EPR: The EPR spectra recorded signals from structural traps, radical centers, and transition metal impurities. The multi-line hyperfine splitting observed in the Mexican sample is characteristic of trace Mn^{2+} ions, typically found as substitutes in carbonate matrices (calcite/dolomite). Conversely, the intense, broad central signal near $g \approx 2$ in the Afghan sample is primarily attributed to S_3^- radical centers, the main chromophore in lazurite. The variations in intensities (Figure 2) and linewidths directly reflect the distinct crystalline environments-carbonate-rich versus silicate-rich-of the analyzed provenances.

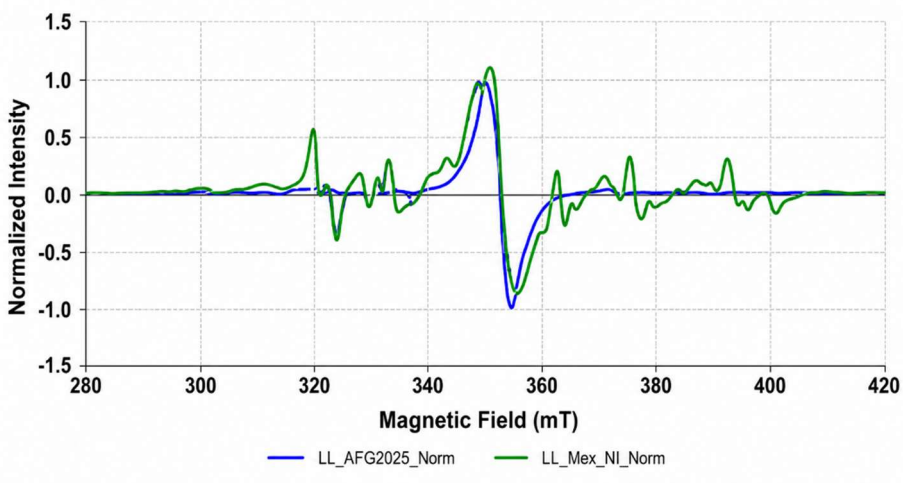


Figure 2 – Normalized EPR spectra of the analyzed samples

4. Conclusion

The combination of XRF, XRD, and EPR techniques proved to be highly efficient in overcoming the difficulty of identifying the provenance of lapis lazuli. The results indicate that the geological environment of Afghanistan samples favors a formation rich in silicates, while the deposit in Mexico generates a matrix rich in carbonates and pyroxenes (diopside) [7, 8]. These structural heterogeneities and the mapped ionic substitutions contribute to the formation of active trapping centers, confirming the potential use of both materials in the development of novel high-dose dosimetric systems.

Acknowledgments

The authors thank the Nuclear and Energy Research Institute (IPEN) for the support and contribution to the development of this work.

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