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Ancient geopolymer in south-American monument. SEM and petrographic evidence

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ABSTRACT

The make-up of the sandstone megalithic blocks, weighing between 130 and 180 tonnes each, from Pumapunku -Tiwanaku, Bolivia, was compared with three geological sandstone sites from the area. The SEM/EDS, XRD and thin section results suggest that the sandstone megalithic blocks consist of sandstone grains from the Kallamarka geological site, cemented with an amorphous ferro-sialate geopolymer matrix formed by human intervention, by the addition of extra alkaline salt (natron) from the Laguna Cachi in the Altiplano, Bolivia.

1. Introduction

Ancient megalithic structure building methods have long been a matter of interest and speculation. Conventional theories suggest that the constituent stone blocks were cut from quarries sometimes remotely located, accurately dressed and lifted into position. For the Egyptian pyramids, MacKenzie *et al.* [1] confirmed in this journal the alternative, but still controversial theory first proposed by Davidovits [2], who suggested that the blocks were a type of early geopolymer concrete. There is currently little research done by materials scientists on these controversial topics. However, from a geopolymer material science point of view, the knowledge that we expect to acquire through this type of archaeological study is manifold. In particular, it generates examples that are useful for the determination of the long-term properties of geopolymer concretes. It helps understanding of the chemical transformation which a geopolymer matrix can undergo over a long time range (hundreds if not thousands of years), and could provide data on the crystallization mechanism and mineralogical evolution.

In this communication we present our preliminary research results on monuments in the South American Andes. They were built 1400 years ago (ca. AD 600) by the Tiwanaku Empire, one of the civilizations of the pre-Columbian Americas (Fig. 1A). The platform on top of the 4 step pyramid of Pumapunku consists of 4 megalithic red sandstone slabs, weighing between 130 and 180 tonnes each (Fig. 1C, D), the largest among the New World monuments. Our study suggests that the slabs are a type of sandstone geopolymer concrete cast on the spot.

One early Spanish conquistador chronicler, Pedro de Cieza de Leon, who visited the Altiplano in 1549, marvelled over the ruins of Pumapunku, wondering what tools could have been used to achieve such perfection (English translation) "*… I asked the natives, whether these edifices were built in the time of the Incas, and they laughed at the question, affirming that they were made before the Incas ever reigned, but that they could not say who made them……*" According to modern archaeology, the monument was destroyed around AD 900, i.e. 500 years before the rise of the Inca Empire.

Travelers mostly agreed that the sandstone was mainly from the Kimsachata mountain range south of Tiwanaku. Yet, it remained unclear how these megaliths were quarried and transported on steep llama tracks as in Fig. 2(1). The first scientific studies conducted and published in the early 1970s by Bolivian archaeologists [3], set out to determine the source of the sandstone employed to construct the Pumapunku complex. They conducted geological studies in 6 drainage valleys, isolating several potential sandstone quarries, totalizing 47 samples. With comparative investigations including X-ray diffraction, XRF, geochemical analysis, and lithic petrography, they concluded that Pumapunku sandstone (20 samples) came from the Kausani *quebrada* (geological site K in Fig. 1B). However, our detailed study of their published chemical analysis contradicts this.

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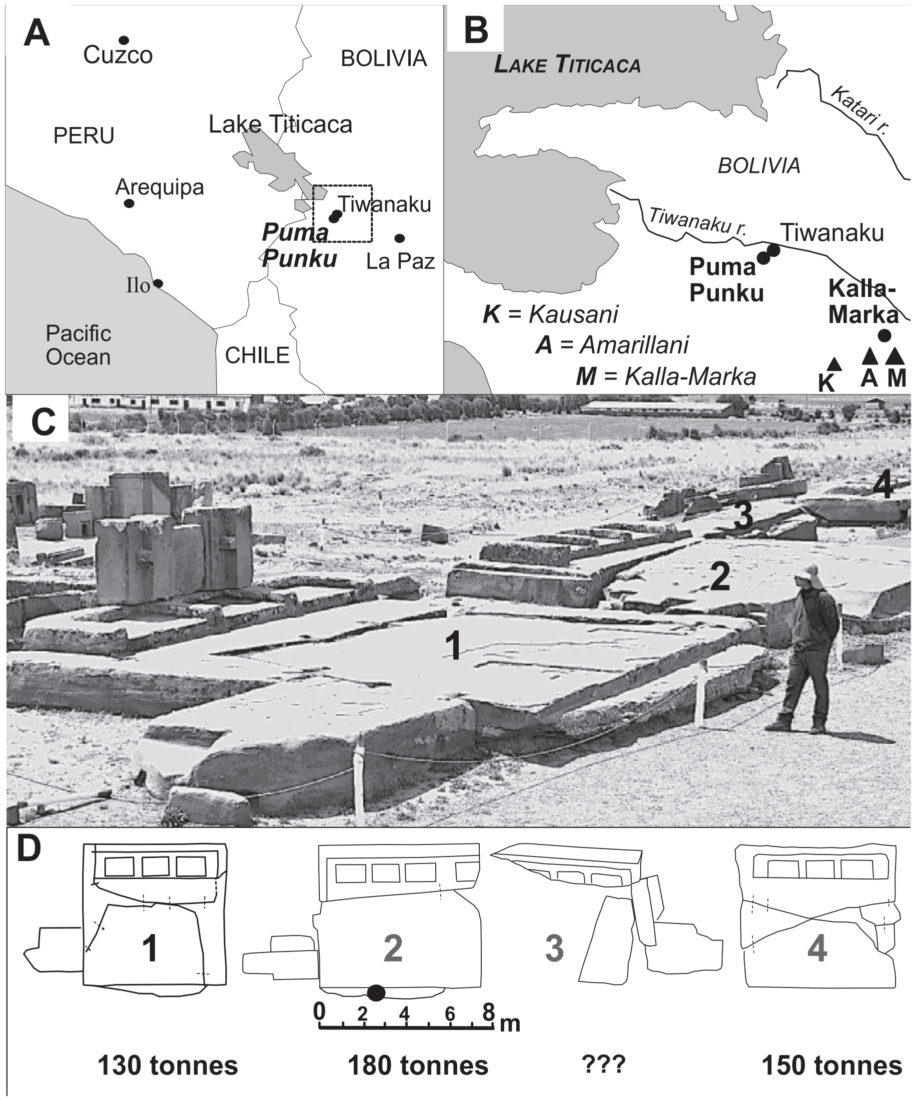

Fig. 1. (A) South American Andes with Pumapunku/Tiwanaku. (B) Location of the geological sites selected in this study. (C) The 4 megalithic red sandstone slabs of Pumapunku. (D) Drawing and dimensions of the 4 slabs.

2. Materials and methods

The Pumapunku red sandstone studied here is from slab No. 2. In Fig. 1D, the sampling place is marked by a black dot. It is near one of the pieces (No. 9) taken and studied in the 1970s. This sample was divided into several fragments for further analysis. For comparison, we selected three geological sites (Fig. 1B): K (Kausani) and A (Amarillani) already studied in the 1970s in which the samples were taken from solid quadratic sandstone blocks as in Fig. 2(2); we added site M located at Kallamarka, a village already known during the ancient time of Tiwanaku. The M sample was not taken from a solid quadratic sandstone block but from a flat weathered sandstone bed as in Fig. 2(4).

The thin 30 μm thick sections were studied under transmitted light (polarized or not) with a Leica 4500 DMP optical microscope. The results are shown in Fig. 2; the thin sections are marked KAU (Kausani), AMA (Amarillani), MAR (Kallamarka) and PP4 (Pumapunku fragment No. 4).

XRD spectra were acquired using a XD8 Advance "BRUKER" AXS (Siemens) spectrometer, calibrated and interpreted according to ICDD/COD international databases from 2013 and DIFFRAC.EVA v.4.1 software. The results are listed in Table 1, with file codes for some elements and semi-quantitative analysis.

XRF data were taken from reference [3], acquired with equipment dating back to 1960.

The SEM images and EDS analysis for Na, Mg, Al, Si, K, Ca and Fe were acquired using a JEOL JSM-6510LV scanning electron microscope. The results are listed in Table 1 and Fig. 3.

3. Results and discussion

In the thin sections of Fig. 2 (optical microscopy), the quartz and feldspar crystal size is: for KAU 100 μm, for AMA 200–400 μm, for MAR and PP4, 150–200 μm (with detrital sandstone fragment particles similar and larger). The mineralogical composition of all samples falls in the range of 40% for quartz, 40% for feldspar and 20% for stone fragments (volcanic and sandstone).

In Table 1, XRD analysis gives the semi-quantitative mineral composition of the sandstone samples. It confirms that the crystalline minerals are mainly quartz and feldspars. Interestingly, we find additional minerals in MAR: calcite $CaCO_3$, kaolinite and illite clays.