THE CHEMISTRY OF NUEVA CADIZ AND ASSOCIATED BEADS: TECHNOLOGY AND PROVENIENCE

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Dating to about 1500-1560, Nueva Cadiz and associated beads comprise the earliest glass bead complex found in the Americas, and many questions regarding their technology and provenience surround them. Analysis of 10 beads from the namesake Nueva Cádiz site in Venezuela and 33 beads collected from an unknown site or sites near Tiahuanaco, Bolivia, provide chemical compositions of their turquoise, dark blue, white, red, and colorless glasses. We analyze the composition of the sand, flux, and colorants that went into their fabrication. The two collections show a common beadmaking tradition and provenience, except for three beads made of high-lime low-alkali (HLLA) glass. Colorants and opacifiers are cobalt for blue, a tin-based agent for white, and copper for turquoise and red. Trace elements associated with cobalt indicate a variable source for this colorant. By comparing the layers of compound beads, we discover technological aspects of bead design and workshop organization. To investigate provenience, we compare the levels of key elements with other glasses of proven origin. There are chemical similarities with glasses made in Venice, identifying it as a candidate to consider when searching for the origin of Nueva Cadiz beads.

INTRODUCTION

Nueva Cadiz and associated beads occur archaeologically from about 1500 to 1560 in regions of Spanish colonial trade from Bolivia to Tennessee. They owe their name to the site in Venezuela where archaeologists first described them. Their place of origin in Europe remains unknown, and some aspects of their technology are unique in the history of beadmaking (Allender 2018; Deagan 1987; Donnan and Stilton 2010; Liu and Harris 1982; Smith and Good 1982). This paper presents an LA-ICP-MS study of beads from the namesake site in Venezuela and an unknown site or sites likely at Tiahuanaco in western Bolivia. After introducing Nueva Cadiz beads, we present the inferred chemical composition of their sand, flux, and colorants, and discuss their fabrication technology and European provenience.

WHAT DO WE KNOW ABOUT NUEVA CADIZ BEADS?

"Nueva Cadiz" refers to drawn tubular beads with a square cross section, found in regions of 16th-century Spanish colonial influence in the Americas. Some are monochrome, but many have three layers of laminated glass. These include Kidd and Kidd (2012) varieties IIIc1-3 and IIIc'4. The latter has a twisted body. In the most widespread varieties, the core is dark blue or gray, the middle layer is white, and the outer layer may be dark blue but often has a characteristic turquoise hue. Size typically varies in the range of 3-10 mm in width and 10-70 mm in length. On some larger specimens, beveled corners reveal the inner layers; this feature is more frequent on more recent examples (Deagan 1987:162-164; Smith and Good 1982). Deagan (1987:163) dates these beads to the first half of the 16th century, and notes their absence at later 16th-century sites. The oldest well-dated examples come from the Nueva Cádiz site in Venezuela, occupied from 1498 to 1543. At present, the youngest tightly dated specimens where we can rule out heirlooms come from the 1559 Tristan de Luna settlement in Pensacola, Florida (John Worth 2021: pers. comm.). These sites frame the circulation of these beads in the Americas between 1500 and 1560.

Nueva Cadiz beads appear in the Americas with other glass beads such as five- and seven-layer chevrons and striped, light gray, olive-shaped "gooseberry" beads. Small dark blue beads that exist in pre-1550 contexts include a ca. 1541-1543 French colony near Québec City (Cooper 2016:262; Delmas 2016:97). The namesake site in Venezuela has square-sectioned monochrome beads that are unknown elsewhere.

Some archaeologists apply the Nueva Cadiz name to square-sectioned tubular beads found on early 17th-century sites in northeastern North America. A style called Nueva Cadiz Twisted – Red Variety (Kidd IIIc'1-3) incorporates a layer of red glass and occurs about 1625-1665 in the French and Dutch colonial trade sphere of New York state and southern Ontario (Bradley 2007:43; Little 2010:224-225; Liu and Harris 1982; Walder et al. 2021). We need further study to understand their relation to archetypal Nueva Cadiz beads.

Smith and Good (1982:1, 46-47) have mapped discoveries in the Americas, but Nueva Cadiz beads have also been excavated in Europe. Divers found 12 production tubes on a 16th-century site in the Venice lagoon (Canal 2013; Zecchin 2005:82-83). In Rouen, a bead and two production tubes came from a ca.1600 beadmaking workshop (Karklins and Bonneau 2019). Antwerp has 30 beads from the house of a 16th-century merchant with ties to Venice (Karklins and Oost 1992). Seville also has one specimen (Deagan 1987:164; Martins Torres 2007:155).

In Portugal, Martins Torres (2007) has inventoried Nueva Cadiz and chevron beads that survive as decorative elements embedded in architectural tiles called *azulejos*. Known examples are in eight buildings from before 1640, notably a chapel at Alcáçovas. At least 30 Nueva Cadiz beads have been recovered from archaeological sites, especially in Lisbon, in contexts from the 16th century, before 1640, and in debris from the 1755 earthquake (cf. Rodrigues 2003, 2007:281-283; Veiga and Figueiredo 2002). Martins Torres also mentions bead collections in Portuguese museums that may include Nueva Cadiz examples.

African varieties tend to differ from their American counterparts. In Angola, archaeologists have reported Nueva Cadiz beads as funerary goods assigned to the 15th or 16th century (Gutierrez 2001:46-50; Gutierrez and Valentin 1995; Rodrigues 1993, 2003:230; 2007:298). The Musée du quai Branly holds 53 examples, about 4 mm wide and long, from Vohémar in Madagascar (inv. no. 71.1961.60.50; Schreurs and Rakotoarisoa 2011). Large type IIIc specimens, 14-20 mm wide from the Lake Chad and Timbuktu regions, likely date to the 19th century (Karklins 2004:43; Liu and Harris 1982:7; Picard and Picard 1993:106).

We find various hypotheses for the place of manufacture of Nueva Cadiz beads. Fairbanks (1968), followed by Smith and Good (1982:12-13), suggested an origin in Andalusia. Karklins and coauthors did not exclude the "tail end" of their production in Rouen or elsewhere in northern France (Karklins and Bonneau 2019; Karklins and Oost 1993:27). Venice is a recurring hypothesis, inspired by its production of similar beads in the last century (Martins Torres 2019:7; Picard and Picard 1993:107; Rodrigues 2007:280, 298; Zecchin 2005:83). As early as 1600, Venetian archives show exports of unspecified bead types to Seville, Lisbon, and Antwerp (Brulez 1965:118, 400, 428). Archaeologists have found Nueva Cadiz production tubes in the Venetian lagoon; however, a cargo of beads likely from Venice, from a 1585 shipwreck at Gnalić, Croatia, has no Nueva Cadiz or chevron styles at all (Delmas 2016:105-106; Jackson 2006:92; Zecchin 2005:82-83). In light of the many finds in Portugal, we may ask whether this country produced Nueva Cadiz and chevron beads. We know that Portugal produced soda glass as early as 1439, but we find no record of its use for beadmaking (Coutinho et al. 2016; Medici 2014:75-79, 108, 507-508). In sum, hypotheses for the origin of Nueva Cadiz beads include Andalusia, Antwerp, northern France, Venice, and Portugal, among others.

PREVIOUS CHEMICAL STUDIES

Lewis (1979) included a colorless square-sectioned bead from the namesake Nueva Cádiz site in the first-ever chemical study of trade beads, but did not comment on the findings. Liu and Harris (1982:8-9) reported another early study that interpreted the presence of soda glass in Nueva Cadiz beads found in Africa and North America, and potash glass in those from Peru. Twenty years later, Portuguese researchers used X-ray fluorescence (XRF) to identify soda glass in all three layers; copper colorant assigned to the turquoise layer and tin opacifier to the white layer (Rodrigues 2003:222-224; Veiga and Figueiredo 2002). They modelled the copper colorant to suggest it derived from chalcanthite ($CuSO_4 \cdot 5H_2O$), a copper sulfate mineral used to color ancient Egyptian faïence (Veiga and Figueiredo 2006).

A subsequent XRF study of beads from a pre-1640 context in Lisbon analyzed six Nueva Cadiz beads, three chevrons, and a blue tubular bead with four red stripes (Rodrigues 2007). This study detected some elements missed previously. Nueva Cadiz and chevron beads contained copper and cobalt colorants, tin opacifier, and lead, while Nueva Cadiz beads also had zinc and high manganese. As for the red-striped bead, its opacifier was antimony, indicating its origin in a different beadmaking tradition or region.

THE PRESENT STUDY

The Venezuela Sample

The first collection in the present study is held by the Florida Museum of Natural History and comes from the site of Nueva Cádiz on Cubagua Island, Venezuela (Figure 1). Christopher Columbus visited the island in 1498 and reported the existence of rich pearl beds. The next year, Spanish

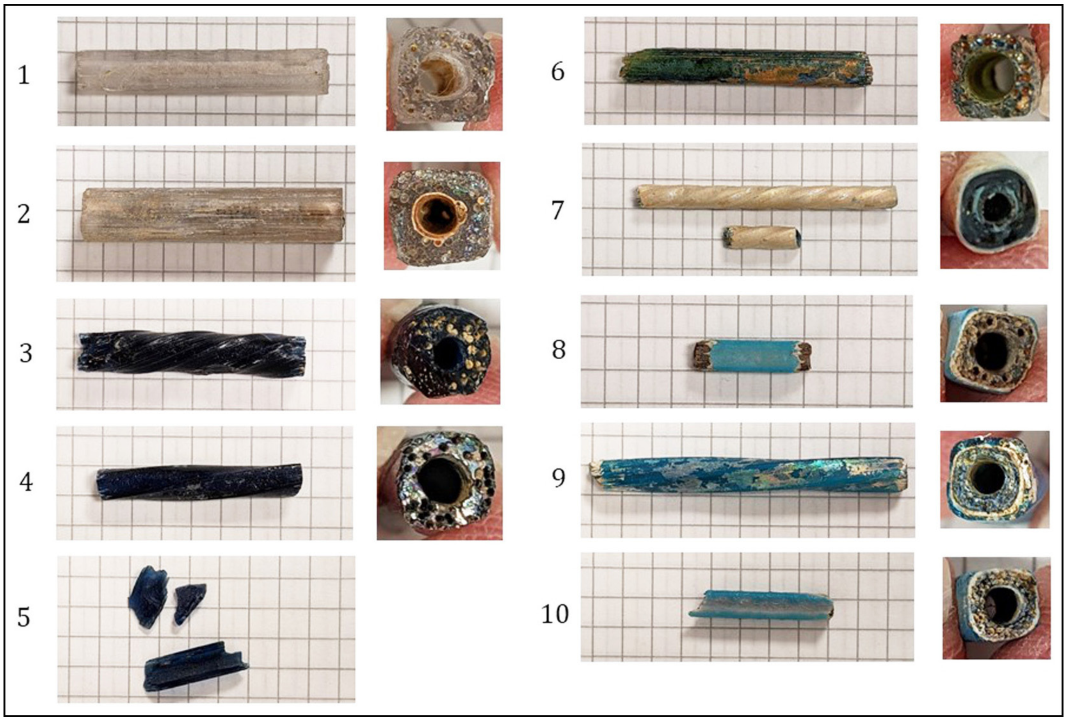

Figure 1. Sampled beads from the Nueva Cádiz site, Venezuela. The grid units are 5 mm (all photos by Brad Loewen unless otherwise noted).

traders acquired 40 kg of pearls from Arawak divers, and settled on the island in 1502. The pearl fishery burgeoned and the settlement expanded to 700 Americans and 223 Europeans by 1527. The pearl beds ran out, however, and the town shrank to 50 residents by 1539. A hurricane destroyed buildings in 1541, and corsairs drove out the last inhabitants in 1543 (Antczak et al. 2019; Romero 2003).

Venezuelan archaeologist Josep María Cruxent excavated the site from 1954 to 1958. Most of the resulting collection resides at the Museo de Nueva Cádiz in La Asunción, but John Goggin, who worked with Cruxent, took some artifacts to the Florida Museum of Natural History and the Yale Peabody Museum. The Florida Museum of Natural History lent 10 beads for this study (Table 1), four of which

Table 1. Bead Samples from the Nueva Cádiz Site, Venezuela.

No.	Length (mm)	Width (mm)	Kidd code	Layer 1 (exterior)	Layer 2	Layer 3	Comments
1	43	8	Ic	Colorless			
2	41	7	Ic	Colorless			
3	37	7	Ic'	Dark Blue			Twisted
4	34	5	Ic13	Dark Blue			Hexagonal section
5	20	5	Ic	Dark Blue			3 fragments
6	42	7	Ic	Greenish			
7	58	4	IIIc'	White	Dark Blue	White	2 fragments; twisted
8	17	6	IIIc	Turquoise	White	Dark Blue	
9	52	6	IIIc'4	Dark Blue	White	Turquoise	Twisted
10	24	4	IIIc	Dark Blue	White	Turquoise	

have three layers, making 18 glass samples in all. These are the oldest archaeologically dated Nueva Cadiz beads known. The associated styles are rare and they shed additional light on the incipient years of the transatlantic bead trade.

The Tiahuanaco Sample

The second bead assemblage lacks an archaeological provenience, but we know part of its history (Figure 2). In 1978, Marvin T. Smith acquired the beads from Liza Wataghani, a dealer in Santa Monica, California, who said they came from Tiahuanaco in western Bolivia. At the time, dealers had only general information on bead provenience, as illustrated by Smith's notes on a different lot: "Excavated in Tiauanaco [sic], but the strings were designed with beads from other sites." While most Nueva Cadiz beads for sale came from Peru, Tiahuanaco was a regular source (Marvin T. Smith 2021: pers. comm.). In 1986, Smith gave the beads to James Bradley, a fellow bead specialist, who transmitted them to Brad Loewen in 2019 for this study.

The 33 beads (Table 2) yielded 72 compositions, three of which turned out to be stone or ceramic (nos. 1, 2, 4). The remainder are typical square-sectioned Nueva Cadiz beads, and are likely more recent than the Venezuela assemblage. They form five groups:

- Group 1 (nos. 3, 5). Two patinated beads, 4.3 and 5.0 mm wide, appear monochrome, but chemical readings show a tin-rich layer sandwiched between two dark blue layers.
- Group 2 (nos. 7, 14, 15). Three monochrome dark blue beads that are 2.7 mm wide and 4-7 mm long exhibit unique bulging sides. The beads have a distinctive high-lime low-alkali (HLLA) composition.
- Group 3 (nos. 6, 8-13, 16-21). Sixteen small beads, about 3 mm in width, have three layers. The outer layer and core are dark blue; the middle layer is white. Due to their small size, only five beads yielded data for all three layers.
- Group 4 (nos. 22-31). The sample includes 10 large beads. With a turquoise outer layer, seven have a dark blue core, three have a core that is blackish, while another two have weakly colored bluish- or greenish-gray cores.
- Group 5 (nos. 32, 33). Two tubular chevron beads with flat ends (IIIp*) exhibit five layers: thin colorless outer layer/white with 10 blue stripes/dark blue/red/colorless core. We did not sample the outermost white and colorless layers.

GLASS ANALYSIS

Methodology

Sampling took place at the Elemental Analysis Facility of the Field Museum in Chicago, using standard procedures for laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) (Dussubieux, Robertshaw, and Glascock 2009). For each sampled glass, we recorded 14 oxides (% of weight) and 43 elements (ppm).

To characterize and compare the base glasses, we calculated the "reduced compositions" that represent their sand and flux components. Following Brill (1999), we included SiO_2, Al_2O_3, and Fe_2O_3 for the sand, and Na_2O, MgO, K_2O, and CaO for the flux. This method eliminates

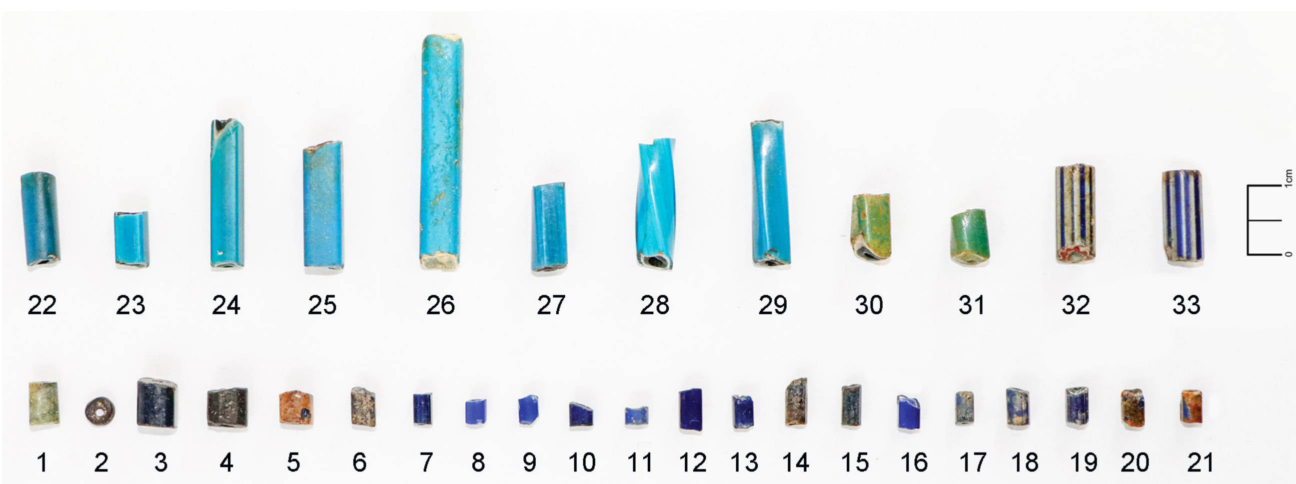

Figure 2. Sampled beads from Tiahuanaco, Bolivia (photo: Saraí Barreiro Argüelles).

Table 2. Bead Samples from Tiahuanaco, Bolivia.

No.	Leng. (mm)	Width (mm)	Kidd code	Grp.	Layer 1 (exterior)	Layer 2	Layer 3	Layer 4	Layer 5
3	7	5.0	IIIc	1	Dark blue	White	Dark blue		
5	4	4.3	IIIc	1	Dark blue*	White*	Dark blue		
6	4	3.0	IIIc	3	Dark blue	White*	Dark blue		
7	4	2.6	Ic	2	Dark blue				
8	3	3.0	IIIc	3	Dark blue	White	Dark blue		
9	3	3.0	IIIc	3	Dark blue	White	Dark blue		
10	3	3.0	IIIc	3	Dark blue	White	Dark blue		
11	3	3.0	IIIc	3	Dark blue*	White	Dark blue		
12	5	3.0	IIIc	3	Dark blue	White	Dark blue		
13	4	3.0	IIIc	3	Dark blue*	White*	Dark blue		
14	7	2.7	Ic	2	Dark blue				
15	6	2.7	Ic	2	Dark blue				
16	4	3.0	IIIc	3	Dark blue	White	Dark blue		
17	4	3.0	IIIc	3	Dark blue	White*	Dark blue*		
18	5	3.0	IIIc	3	Dark blue*	White	Dark blue		
19	4	3.0	IIIc	3	Dark blue	White*	Dark blue*		
20	4	3.0	IIIc	3	Dark blue*	White*	Dark blue		
21	4	3.0	IIIc	3	Dark blue*	White*	Dark blue		
22	13	4.5	IIIc	4	Turquoise	White	Dark blue		
23	8	4.0	IIIc	4	Turquoise	White	Dark blue		
24	22	4.0	IIIc	4	Turquoise	White	Dark blue		
25	18	5.1	IIIc	4	Turquoise	White	Dark blue		
26	34	5.1	IIIc	4	Turquoise	White	Bluish		
27	13	4.2	IIIc'4	4	Turquoise	White	Dark blue		
28	21	4.5	IIIc'4	4	Turquoise	White	Dark blue		
29	21	4.5	IIIc'4	4	Turquoise	White	Dark		
30	9	4.8	IIIc'4	4	Turquoise	White	Dark blue		
31	8	4.7	IIIc'	4	Turquoise	White	Greenish		
32	14	5.2	IIIp	5	Colorless	White w/ 10 blue stripes	Red	White	Colorless
33	14	5.2	IIIp	5	Colorless	White w/ 10 blue stripes	Red	White	Colorless

* Non-sampled glass.

the dilution caused by colorants and opacifiers that can account for 25%-28% of glass by weight.

We used the concept of *chaîne opératoire* as a bridge to link chemical compositions to beadmaking technology. The *chaîne opératoire* represents beadmaking as a sequence of steps, in a thought process that is familiar to archaeologists. It conceptualizes artifacts as the fruit of a chain of operations, meaning that beadmakers introduced different chemical components into the glass material at specific steps or operations. By identifying sets of elements and associating them with specific operations, we can reconstruct aspects of workshop organization. This concept is inherent in the creation of chemical subsets such as reduced composition, and in the definition of various units such as glass batches, color lots, glass layers, and bead groups within a site. Each of these subsets and units corresponds to a step in the *chaîne opératoire*.

To study provenience, we compared Nueva Cadiz and associated beads with other glasses that have a proven provenience. We focused on elements used to this end by other researchers, namely potash, alumina, titanium, zirconium, hafnium, and neodymium. When comparing elements reported in ppm with oxides in % wt, we used standard stoichiometric conversion values (e.g., TiO_2 % wt / Ti ppm = 1.6682).

Global Glass Composition

Reduced compositions for the study collections show two glass types: soda-lime for the majority of samples and high-lime low-alkali (HLLA) for three samples (Tables 3 and 4).

Soda-Lime Glasses

All the glasses from Venezuela and most of those from Tiahuanaco have a soda-lime composition. The most abundant oxides after silica are soda (11.0%-15.6%) and lime (4.9%-10.3%). The combination of high soda concentrations with potash and magnesia above 1.5% suggest the use of the ashes of halophytic plants that grow in salty soils around the

Table 3. Average Reduced Compositions for Glass Colors from Venezuela.

	Colorless	Dark blue	Greenish	White/blue/white		Nueva Cadiz (3)		
				Blue	White	Dark blue	White	Turquoise
n=	2	3	1	1	2	3	3	3
SiO_2	75.4%	71.6%	66.7%	71.9%	69.1%	73.2%	71.8%	71.2%
	0.4%	0.2%			0.4%	4.5%	2.5%	3.1%
Na_2O	12.9%	16.7%	15.6%	11.0%	11.8%	12.5%	13.0%	13.7%
	0.3%	0.3%			0.2%	0.2%	1.2%	0.8%
MgO	2.5%	1.5%	1.8%	2.8%	3.4%	2.5%	3.1%	3.1%
	0.1%	0.0%			0.1%	0.6%	0.5%	0.9%
Al_2O_3	0.6%	1.0%	1.2%	0.9%	1.6%	0.9%	1.0%	1.1%
	0.0%	0.0%			0.2%	0.2%	0.2%	0.2%
K_2O	2.7%	3.1%	2.9%	4.4%	4.2%	3.6%	3.3%	2.9%
	0.1%	0.1%			0.1%	0.6%	1.2%	1.1%
CaO	5.7%	4.9%	7.3%	8.6%	9.2%	6.4%	7.2%	7.6%
	0.0%	0.2%			0.9%	2.5%	1.9%	2.1%
Fe_2O_3	0.2%	1.1%	4.5%	0.4%	0.7%	0.9%	0.6%	0.4%
	0.0%	0.3%			0.1%	0.8%	0.3%	0.1%

Standard deviations are in the white cells, when there was more than one analyzed sample.

Table 4. Average Reduced Compositions for Glass Colors from Tiahuanaco.

	HLLA	Small beads (18)		Large Nueva Cadiz beads (10)						Chevron beads (2)			
	Blue	Blue	White	Bluish	Greenish	Dark	Blue	White	Turquoise	Colorless	Blue	Red	White
n=	3	23	7	1	1	1	7	10	10	4	2	2	2
SiO_2	65.8%	68.8%	69.4%	72.9%	69.6%	66.7%	69.3%	70.2%	70.6%	68.5%	69.8%	65.4%	68.8%
	2.4%	1.5%	0.9%				1.0%	1.7%	1.7%	0.5%	0.5%	0.7%	0.7%
Na_2O	4.5%	12.2%	13.6%	11.8%	13.8%	13.7%	13.6%	13.6%	13.4%	14.3%	13.0%	13.6%	13.9%
	1.8%	2.3%	0.7%				1.1%	1.0%	1.3%	1.2%	1.1%	1.1%	1.6%
MgO	2.7%	3.2%	3.3%	2.0%	3.2%	4.1%	2.8%	3.0%	2.9%	3.3%	3.2%	3.1%	3.3%
	0.4%	0.2%	0.3%				0.2%	0.5%	0.6%	0.2%	0.1%	0.0%	0.0%
Al_2O_3	3.4%	1.4%	1.0%	0.8%	1.1%	1.7%	0.9%	1.0%	1.0%	1.1%	1.3%	1.3%	1.3%
	0.8%	0.9%	0.1%				0.2%	0.2%	0.3%	0.2%	0.2%	0.4%	0.1%
K_2O	3.5%	2.6%	2.5%	5.4%	5.2%	3.1%	3.2%	3.7%	3.7%	2.2%	2.6%	2.6%	2.5%
	0.6%	0.4%	0.1%				1.0%	1.4%	1.5%	0.5%	0.5%	0.8%	0.7%
CaO	18.3%	10.3%	9.8%	6.8%	6.6%	9.9%	7.6%	8.1%	7.8%	9.9%	9.0%	8.8%	9.5%
	3.5%	2.0%	0.8%				1.2%	1.4%	1.4%	0.1%	0.7%	0.1%	0.1%
Fe_2O_3	1.7%	1.6%	0.5%	0.3%	0.5%	0.8%	2.4%	0.5%	0.6%	0.6%	1.1%	5.2%	0.7%
	0.5%	0.5%	0.1%				1.6%	0.1%	0.3%	0.2%	0.1%	0.6%	0.3%

Standard deviations are in the white cells, when there was more than one analyzed sample.

Mediterranean (Sayre and Smith 1961). The pale bluish and greenish glasses from Tiahuanaco have lower soda and lower lime but higher potash, but we note that only one sample of each color was analyzed. Alumina below 2% indicates access to a rather pure source of silica (Cagno et al. 2012).

HLLA Glasses

Three small dark blue beads from Tiahuanaco contain a different glass type characterized by high lime (CaO) averaging 18.3% and low alkali (Na_2O + K_2O) totaling only 8.0% in reduced composition. Alumina at 3.4% is higher than in the soda-lime glasses. The combination of high lime and alkali below 10% defines "high-lime low-alkali" (HLLA) glass (Dungworth and Cromwell 2006). The beads containing this glass are also visually distinct, being the only monochrome specimens from Tiahuanaco, and having a smaller section (nos. 7, 14, 15). They exhibit bulged sides, a feature not seen in the other square-sectioned beads (Figure 3). The HLLA glasses also stand out for their high phosphorus oxide concentrations (P_2O_5) in the range of 1.5%-2.0%, compared to other samples at 0.1%-0.5%. According to Stern (2017), phosphorus content of 0.2%-1% indicates the use of soda plants to make the flux, while 1%-3% identifies wood ash. Two of the HLLA beads (nos. 7, 14) have low strontium (185 and 275 ppm), about half the average for soda-lime glasses (508 ppm), also denoting a different flux material (Degryse and Shortland 2020; Dungworth 2013; Dungworth, Degryse, and Schneider 2009).

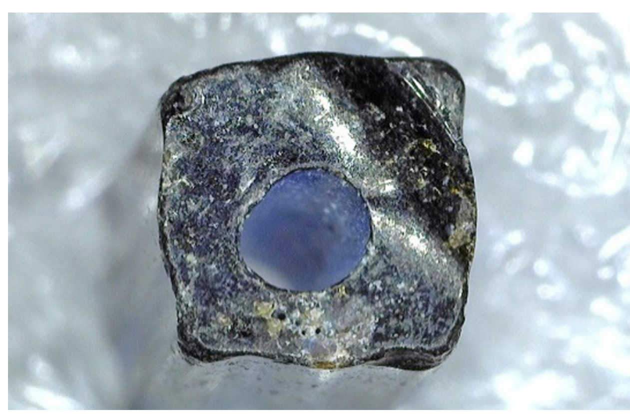

Figure 3. Tiahuanaco blue HLLA bead no. 7 with atypical bulging sides; 2.7 mm wide.

Usually found in bottles, HLLA glass is a sub-type of potash glass that appeared in Germany, and spread to northern France and England in the 16th century (Historic England 2018; Mortimer 1995; Schalm et al. 2007). In these regions, noble families controlled the production of potash-glass windowpane, while commoners made HLLA bottles. To reinforce these social distinctions, some glassworks had separate furnaces for these glasses (Dungworth and Cromwell 2006:162; Klaës 2021). We find few examples of HLLA glass in southern Europe. Researchers have reported isolated artifacts in Altare and Portugal, but none in two large Venetian assemblages (Cagno et al. 2012; Jackson 2006; Medici 2014:418-420; Palamara et al. 2017). The HLLA beads appear to show a northern European influence in the Tiahuanaco assemblage.

Sand Composition

Silica (SiO_2) is the major constituent of glass. Quartz sand consists almost entirely of silica, but it also contains other elements that enter glass involuntarily – principally aluminum and iron in the form of Al_2O_3 and Fe_2O_3. Ratios of silica, alumina, and iron allow us to characterize the sand that went into the Venezuela and Tiahuanaco beads.

The sand used for the Venezuela glasses shows a high silica content: 96.2%-98.6% of a hypothetical sand containing only SiO_2, Al_2O_3, and Fe_2O_3. Accordingly, these glasses have low levels of impurities. High iron in the greenish bead, no doubt added voluntarily, explains its tint. Aluminum levels are low (0.6%-1.6%). Slightly higher iron (0.7%-1.1%) in dark blue and white glasses may reflect coloring and opacifying additives.

The Tiahuanaco beads (excepting the HLLA beads) also have low levels of sand contaminants. The sand used to make the white and turquoise layers in Nueva Cadiz beads has silica purity attaining 97.7%. Aluminum is generally low (0.8%-1.7%), especially in Nueva Cadiz beads. Iron is slightly elevated (1.1%-2.4%) in dark blue glasses, and particularly in the red glass of chevron beads (5.2%). Iron above 0.8% probably results from coloring processes (Jackson 2005). When we account for this added iron, we estimate the silica purity of sand in Tiahuanaco soda glass beads at ~97% SiO_2, and only ~91% in HLLA beads.

In both collections, Nueva Cadiz beads have low aluminum levels in all glass colors (0.9%-1.1%), consistent with a source of very pure sand. Iron is consistently very low in the white and turquoise layers (0.1%-0.2%), but some differences appear in the dark blue layer. Fe_2O_3 is moderately high in Venezuela dark blues, and very high in Tiahuanaco samples. The standard deviation for Fe_2O_3 in dark blue is high in Nueva Cadiz beads, indicating wide variations among beads. This variability likely betrays a diversity of coloring recipes, and not different sand sources.

In the Italian tradition of soda glassmaking that spread through much of Europe, artisans accorded great value to sand purity. Venetian glassmakers preferred crushed river cobbles to make *cristallo*, the clearest soda glass attainable in the 15th-17th centuries, which shows 97%-99% silica in sand (Janssens et al. 2013). Glass beads, despite their exuberant palette of colors, often contain similarly pure sand, a feature that identifies beadmaking as a subsidiary of the soda-glass industry on which it relied for base glass.

Flux Composition

In soda-lime glass (soda glass) of the 15th-18th centuries, plant ash had both a fluxing and a stabilizing function. The ash usually derived from sodic plants that thrive in saline soils on the Mediterranean coast. Syria and Spain were major producers and exporters. Syria sold its soda to Venice, while Alicante shipped its barilla to glassmakers throughout Western Europe (Ashtor and Cevidalli 1983; Girón-Pascual 2018; Jacoby 1993; Verità 2021).

While soda glass comprises most trade beads, it co-existed with an array of glass types in Europe in the 15th-18th centuries. Gratuze and Janssens (2004:672) developed a ternary graph to sort glasses by flux type using three-way ratios of CaO, Na_2O, and K_2O+MgO, which are the principal flux components in glass. Four major glass types fall in different areas of the graph: 1) natron glass from the Roman period, 2) soda glass from medieval and early modern Europe, 3) mixed-alkali glass from northern France in the 16th-18th centuries, and 4) potash or "forest" glass from northern Europe in medieval and modern times. Our review of published data finds that 95% of analyzed beads from 1580-1780 fall in the soda-glass area (Figure 4, area 2), generally in its "lower" half where Na_2O contributes 30%-50% of the principal flux components. The remaining analyzed beads contain potash or mixed-alkali flux, or they consist of lead glass; these glasses occur in beads made after ca.1670.

The Venezuela beads are made of soda glass, with relatively high Na_2O (40%-64%) compared to published compositions (Figure 4). We see that the samples form several clusters of two or three similar glasses. In fact, each Nueva Cadiz bead forms a cluster to itself. All the colors of a bead have near-identical flux compositions, but each Nueva Cadiz bead is distinct from the others. The different colored glasses in a bead may derive from a single batch of base glass, but no two Nueva Cadiz beads come from the same batch. The two colorless beads likely came from the same glass batch, as did the three dark blue specimens.

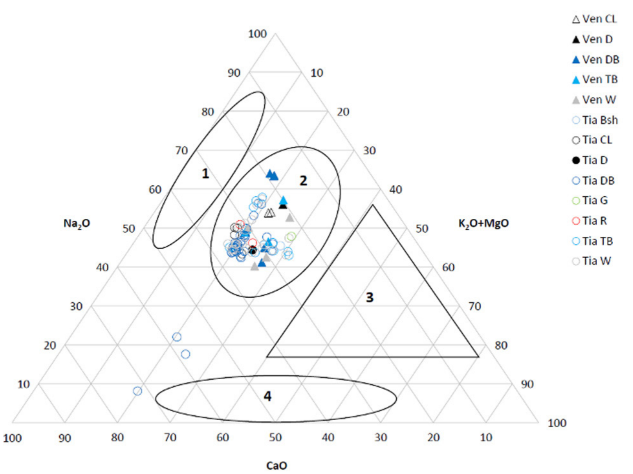

Figure 4. Ternary graph of flux compositions, following the glass typology of Gratuze and Janssens (2004): 1) Roman natron glass; 2) soda-lime glass, 14th-18th centuries; 3) medieval and post-medieval mixed-alkali glass; and 4) medieval and post-medieval potash or "forest" glass (all graphics by Laure Dussubieux).

The Tiahuanaco glasses also fall in the soda-flux area of the ternary graph, except for three HLLA glasses. The close clustering of most samples indicates the use of a homogeneous plant ash (Figure 5). Each Nueva Cadiz bead shows nearly identical flux composition in all three colored layers, indicating the use of a single base glass batch to make all the colors. Three beads may come from the same glass batch (nos. 23, 24, 28) and three other beads from another batch (nos. 25, 27, 30), but each remaining Nueva Cadiz bead comes from its own glass batch.

Figure 5. Detail showing dispersed pairs and triplets of glasses in beads from Venezuela, tight clustering of dark blue and white glasses in small beads from Tiahuanaco, and the wider distribution of glasses in Nueva Cadiz beads from Tiahuanaco.

Of the 15 small three-layer beads from Tiahuanaco, the majority likely emanate from a single glass batch, and the others show only slight differences (Figure 6). Remarkably, beads from the same glass batches stayed together as lots until our time.

Colorant and Opacifier Compositions

Most of the glasses in this study are dark blue, white, or turquoise, but colorless, red, bluish gray, greenish gray, and blackish glasses are also present. Reduced compositions show no significant differences of base glass among colors.

Dark Blue Glass

We recorded seven compositions of dark blue glass from Venezuela and 33 from Tiahuanaco.They include the three monochrome HLLA beads from Tiahuanaco. Most dark blue glasses form the inner and outer layers of small Nueva Cadiz beads from Tiahuanaco (n=21), while 10 samples form the inner layer of large Nueva Cadiz beads. Other dark blue samples, all from Venezuela, come from three large monochrome beads and the white/blue/white bead.

The main coloring ingredient is cobalt that imparts a deep blue when present in a few hundred to a few thousand ppm. Cobalt is also a source of information on beadmakers' supply networks, as cobalt ore contains additional elements that help determine its provenience. Gratuze et al. (1996) show an evolution of ores used to color European glass from Roman times to the 18th century. The sequence culminates with ore from the Schneeberg mine in the Erzgebirge region of Germany. This ore has higher Ni, As, and Bi that go hand in hand with higher cobalt. Exceptionally, cobalt pigments found in majolica glaze made in Aragon show other ore profiles with higher Cu or Mn, possibly from Pyrenean mines (Pérez-Arantegui et al. 2009).

Two Venezuela beads feature the Co-Ni-As-Bi profile associated with Schneeberg ores. The other Venezuela beads show a different profile, with low As and Bi and only Ni in higher concentration. We may infer that cobalt ores used to color these beads came from different sources (Figure 7).

In the Tiahuanaco beads, higher Co, Ni, As, and Bi in the dark blue soda-lime glasses all match the Schneeberg profile. These glasses also have higher iron and manganese concentrations than other glass colors. Since cobalt often occurs with iron and manganese in nature, it can bring these elements into glass involuntarily (Dehaine et al. 2021; Gratuze, Pactat, and Schibille 2018). The specific cobalt ore may explain MnO values above 0.6% and Fe_2O_3 above 1.0% in dark blue glasses. The three HLLA beads, however, reveal

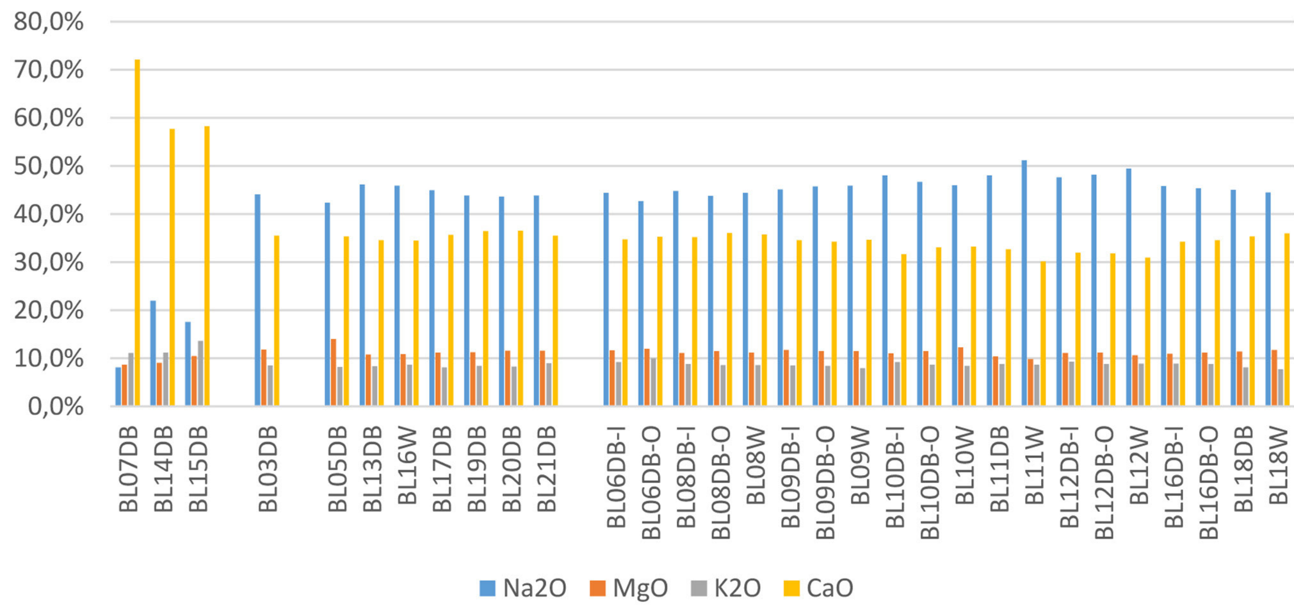

Figure 6. Principal flux components in glasses of small beads from Tiahuanaco, showing their ratios as % of their total. Most samples have near-identical flux compositions. DB = dark blue, W = white, I = inner layer, O = outer layer.

three different cobalt-related profiles. No. 7 has very low As, Bi, and Ni; no. 14 has very high As but low Ni and Bi; and no. 15 has low levels of all three elements. We suggest the use of three different cobalt ores for coloring the HLLA glasses, showing a diversity of cobalt sources in contrast to the soda-lime beads.

Most dark blue glasses contain traces of copper, usually below 0.6%, which are also compatible with impurities in cobalt ore (Figure 8). Copper in blue glass, however, attains 1.2%-3.8% in five Nueva Cadiz beads, one from Venezuela (no. 9) and four from Tiahuanaco (nos. 23, 24, 28, 30). We notice similar levels in turquoise glasses (2.0%-3.6%) where copper is the main coloring agent. Possibly, the beadmakers converted surplus turquoise stock into these copper-rich dark blue glasses, by adding cobalt colorant and tin-lead opacifier.

The majority of dark blue glasses also contain significant levels of tin and lead, as much as 9% of total glass composition (Figure 9). These elements partially opacify the glass and make it darker, as less light passes through. In white glass, these elements constitute the dominant opacifier and colorant. Their average level in dark blue glass (7.5%) is about 30% of that in white glass (25%). Since we have no previous layer-by-layer LA-ICP-MS studies of compound beads, or of the tin-lead combination itself, we considered whether these elements could have diffused from nearby white glass during the beads' fabrication or lifespan, or represent involuntary contamination during sampling.

We find, however, that there are similar levels of tin and lead in monochrome dark blue beads that have no white glass as a possible source of diffusion or contamination. The presence of tin and lead in dark blue glass was either purposeful to create opacity or resulted from recycling previously opacified glasses.

We believe these elements had a purpose because of a pattern seen in the small three-layer beads from Tiahuanaco (Figure 10). In these beads, the dark blue core has moderate tin and lead (4%-9%), whereas the outer dark blue layer has low levels (0.6%-2%). As well, the tin-rich core has low cobalt (820-3035 ppm), contrary to the outer layer that has lower tin and high cobalt (3296-6065 ppm Co). In the outer layer, high cobalt combined with low opacifier produced an intense, diaphanous blue that allows light to enter and reflect back from the middle white layer. A glassmaking treatise describes an analogous effect of tin in the manufacture of mirrors: "It is not the glass that makes the mirror, but the tin; because without the tin, it would be impossible to reflect objects held up to it" (Haudicquer de Blancourt 1718, 2:242). The judicious dosage of cobalt and opacifier in each bead layer similarly used tin to reflect light and create a shimmering effect.

White Glass

We analyzed 5 samples of white glass from Venezuela and 19 from Tiahuanaco. Most form the middle layer of small and large Nueva Cadiz beads (n=20). Two samples

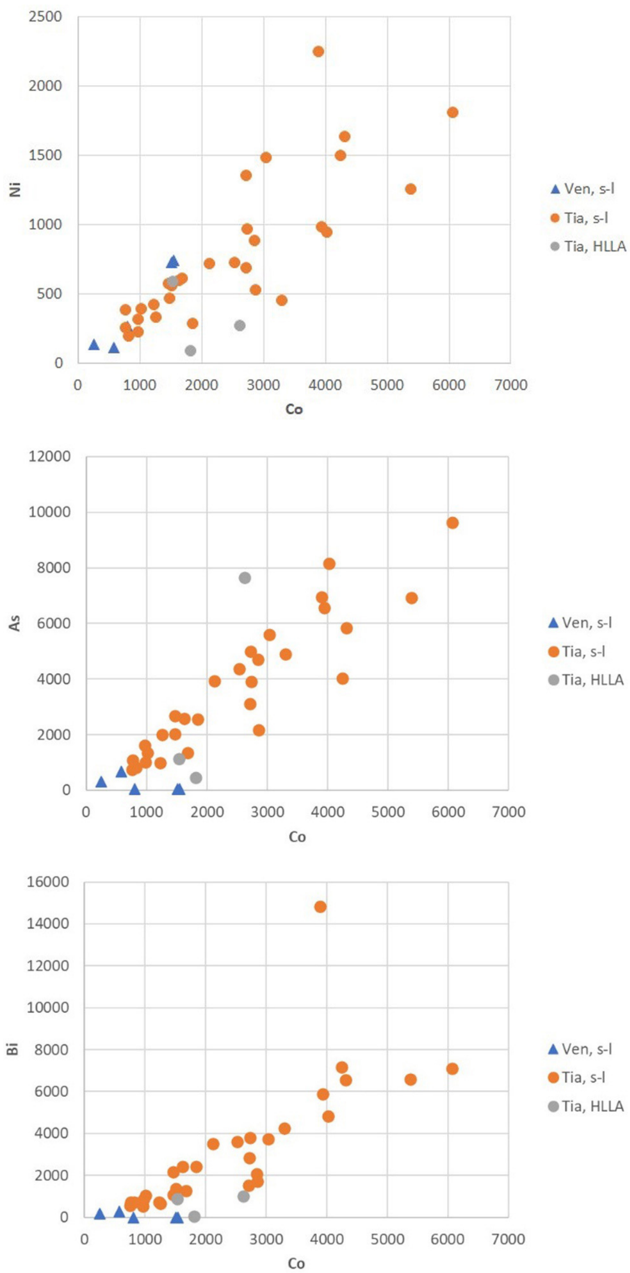

Figure 7. Cobalt ratios to nickel, arsenic, and bismuth in dark blue glasses. The outlier with high nickel and bismuth is Nueva Cadiz bead no. 25.

come from the white/blue/white Venezuela bead and two from the Tiahuanaco chevrons. All the white glasses contain both tin and lead that typically comprise 22%-28% of the glass matrix. The ratios of SnO_2 to PbO show three recipes ranging from 6:10 to 9:10 by weight (Figure 11). Higher indium (In) in these samples is typical of many tin ores (Benzaazoua et al. 2003; Comendador Rey et al. 2017; Lerouge et al. 2017; Wang et al. 2016).

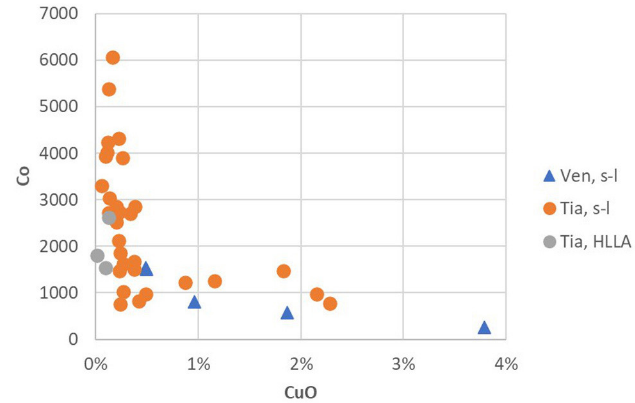

Figure 8. Cobalt and copper (CuO) levels in dark blue glasses. Copper levels below 0.6% are consistent with cobalt ore.

Tin as SnO_2 (cassiterite) forms white crystals that produce an opaque white aspect when dispersed in colorless glass (e.g., Matin 2019; Tite, Pradell, and Shortland 2008). Lead decreases the solubility of cassiterite in glass, thus favoring its crystallization (Molera et al. 1999). Starting in the early 15th century, Venetian glass recipes describe the creation of opaque white glass called *lattimo* (e.g., Moretti, Salerno, and Tommasi-Ferroni 2004; Verità and Zecchin 2009). Artisans made a white opacifier by calcinating metallic lead and tin, to make a white powder called calx. They mixed this powder into molten glass to impart an opaque white hue (Billeck and McCabe 2018; Matin 2019). Trade beads found in North America show a chronology of tin use for opacifying. Only tin was used before 1625, after which antimony appeared and soon became the exclusive opacifier. The tin-antimony shift happened ca. 1625-1650 in Dutch beads, and ca. 1650-1675 in French beads. Lead also vanishes from trade beads at this time, except for rare lead glasses, and yellow or amber colorants. Arsenic is the opacifier in beads from the late 18th and 19th centuries (Hancock 2013).

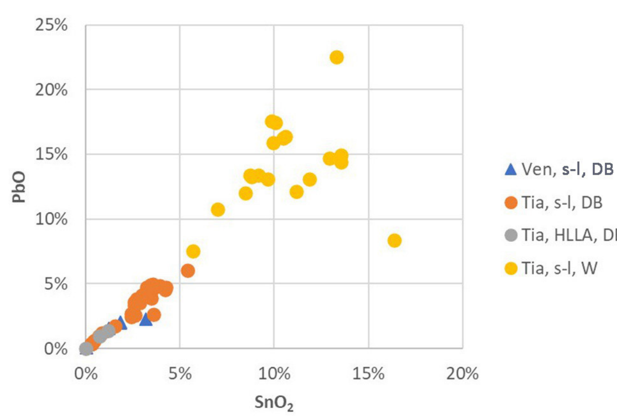

Figure 9. Tin and lead content, showing their consistent ratio in all glass colors.

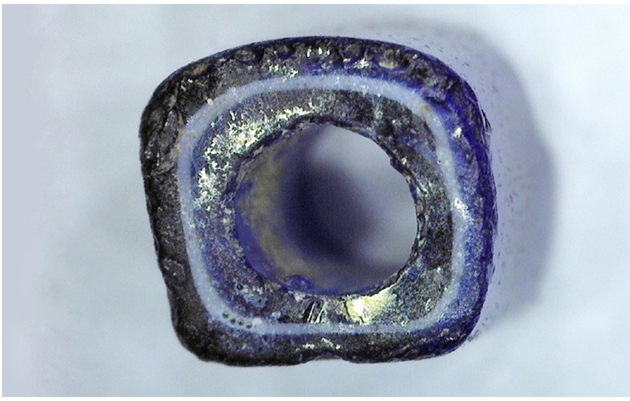

Figure 10. Tiahuanaco small bead no. 8.

Beadmakers used calx not only to create white glass, but also to slightly opacify other colors. Thus, bead layers have stepped levels of tin and lead. The opaque white middle layer has 25% on average, while the dark blue core has 7.5%. As for the outer layer, the small dark blue beads and five large turquoise examples have 0.6%-2.2% tin and lead, while eight turquoise glasses have insignificant levels of opacifier (Figure 9).

Turquoise Glass

We measured 3 turquoise glasses from Venezuela and 10 from Tiahuanaco, all from the outer layer of Nueva Cadiz beads. The turquoise color derives from copper in the form of Cu^{2+} that develops in a normal atmosphere requiring little technical expertise. Calculated as CuO, copper concentrations range from 3.0%-3.6% in the Venezuela samples and 2.0%-3.4% in those from Tiahuanaco. Much less copper can still produce a vibrant turquoise color in glass. In three beads from Tiahuanaco (nos. 22, 25, 31), the turquoise layers have tin and lead combining for 1.4%-2.3% (Figure 12). This level is similar to the outer dark blue layer of small Nueva Cadiz beads (≤ 2%), indicating a similar approach to adjusting the amount of light passing through the beads' outer layer (Figure 13). The compositions of these layers illustrate the beadmakers' use of opacifier levels to create different light effects.

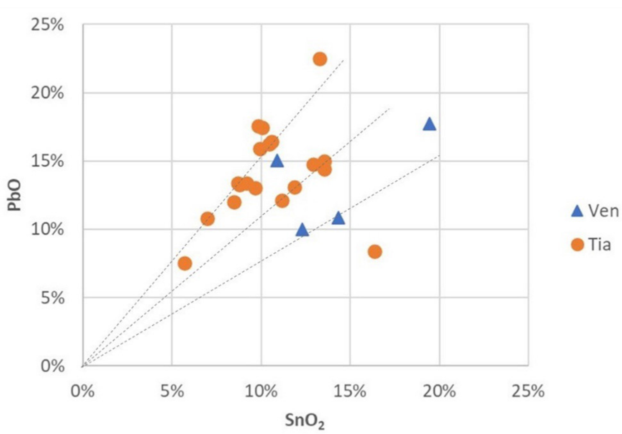

Figure 11. Tin and lead levels in the white glasses of small Nueva Cadiz and tubular chevron beads. The Tiahuanaco outlier (bottom right) is from a chevron bead. Diagonal lines show similar ratios of tin and lead in several beads.

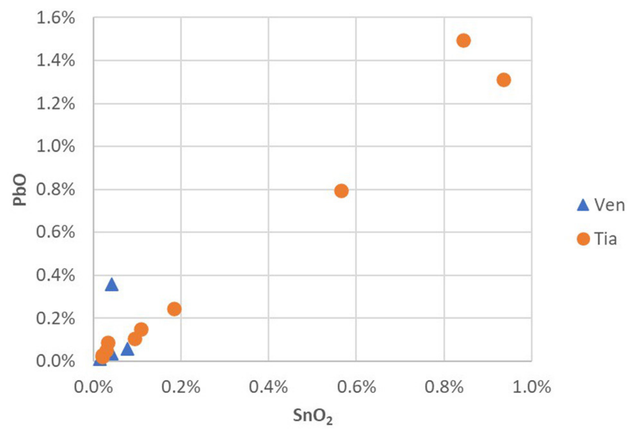

Figure 12. Tin and lead levels in turquoise glasses of Nueva Cadiz beads. Combined levels below 0.4% are the norm.

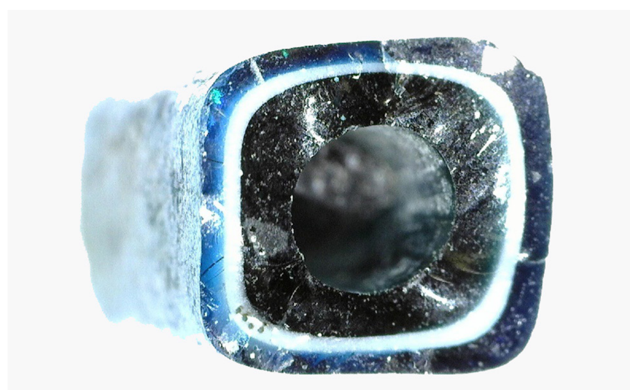

Figure 13. Nueva Cadiz bead no. 23 from Tiahuanaco.

Red Glass

We obtained two compositions of the red glass in the chevron beads from Tiahuanaco (Figure 14). Red is a color usually produced by the addition of copper either as metal scraps or as a prepared oxide. The red glass samples contain moderate amounts of copper (CuO = 1.0% and 1.6%). To obtain red, glassmakers needed to skillfully maintain a reducing atmosphere (depleted of oxygen) in the furnace. Iron found in significant concentrations (Fe_2O_3 = 4.6% and 5.4%) may have acted as an internal reducer that facilitated the precipitation of copper as metallic copper or cuprous

Figure 14. Five-layer chevron bead no. 32 from Tiahuanaco.

oxide crystals, which produce an opaque red color (Ahmed, Ashour, and El-Shamy 1977). "Of all colored glasses," Cannella (2006:171) states, "red glass certainly gave the most trouble to master glassmakers over the centuries." She cites recipes for red glass that used cuprous and ferrous ingredients variously described as kettle offcuts, iron filings, Saffron of Mars, Saffron of Iron, magnesium iron, and carbon-rich particles of iron slag that accumulated around a blacksmith's anvil.

Colorless Glass

We sampled four colorless glasses: two colorless Nueva Cadiz beads from Venezuela and the colorless cores of two chevron beads from Tiahuanaco, one of which we sampled three times (no. 33). Glass has a natural bluish, greenish, or brownish tint due to the presence of iron in silica sand. Glassmakers had various ways of minimizing the intensity of this tint. They could choose a sand with the least amount of iron possible, they could control the atmosphere in the furnace to produce an iron species with the least tinting power, or they could add a decoloring element such as antimony, arsenic, or manganese to neutralize the ferrous tint (Meulebroeck et al. 2010).

Iron levels (measured as Fe_2O_3) are 0.2% in the colorless beads from Venezuela, and 0.4%-0.9% in the Tiahuanaco glasses. While these levels are among the lowest of all color categories, they are similar to those in white (0.3%-0.7%) and turquoise glasses from Tiahuanaco (0.3%-1.1%). We note, however, that the sample size for colorless glass is relatively small.

We may ask whether a decoloring agent such as antimony, arsenic, or manganese produced the colorless aspect. Antimony (Sb) does not rise above a few tens of ppm in any of our glasses, and colorless glasses show no enrichment. Arsenic (As) is 3-4 ppm in colorless beads from Venezuela and 110-147 ppm in Tiahuanaco colorless glasses. This concentration is below the few hundred ppm in turquoise where arsenic enters as a copper impurity, and the few thousand ppm in dark blue where it is an impurity of cobalt.

As for manganese (MnO), it occurs at 0.3%-0.8% in the colorless glasses (Figure 15). Manganese has several possible pathways into glass, and its interpretation is complex. Soda plant ash can contribute ca. 0.02%-0.06% (Barkoudah and Henderson 2006; Occari, Freestone, and Fenwick 2021; Phelps et al. 2016; Schibille, Sterrett-Krause, and Freestone 2017). As a sand impurity, it can enter glass at levels below about 1%. Used as a colorant, manganese can create a spectrum of pink and purple hues, culminating with black when present at concentrations higher than about 3% (Hancock 2013). Finally, in its role as "glassmaker's soap," manganese can eliminate ferrous tints at concentrations of 1%-2%, if iron is found at similar levels in colorless glass (Jackson 2005, 2006:88; Sayre 1963). In our colorless glasses, MnO and Fe_2O_3 (0.2%-0.9%) fail to cross the threshold of the decolorizing hypothesis. In fact, manganese levels in colorless glass are no higher than in any colored glass, so we cannot infer its addition with the aim of washing a ferrous tint out of a glass batch. Thus, the colorless aspect of these glasses derives from the use of high-purity sand and a proficient control of furnace conditions.

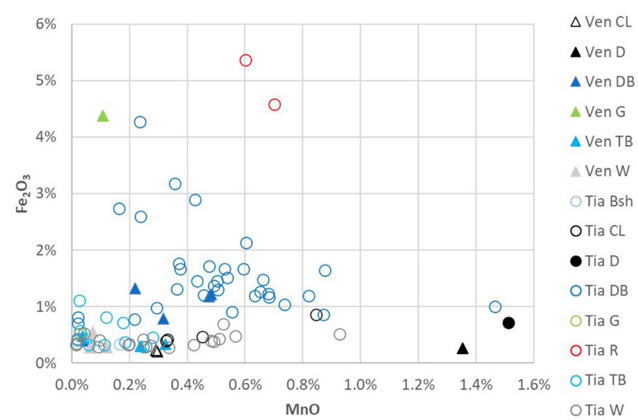

Figure 15. Manganese relative to iron is no richer in colorless than in colored glasses, so it did not serve as a decolorant.

Distinctive Inner Layer Colors of Nueva Cadiz Beads

In three Nueva Cadiz beads from Tiahuanaco, the core has a different color (Figure 16). Two with weak "bluish" and "greenish" tints (nos. 26, 31) have lower soda and lime but higher potash, and show no added colorant. The third has a "dark" blackish gray color (no. 29). It has higher lime and its color derives from added manganese (1.5%).

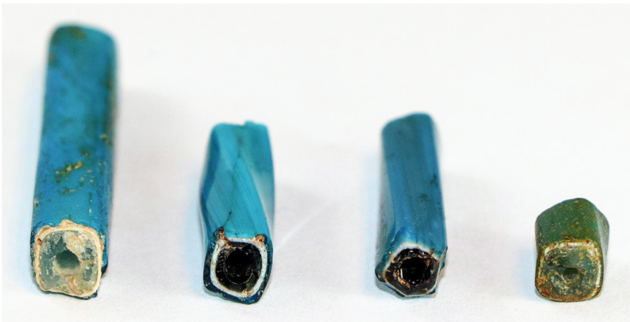

Figure 16. Color variants in the cores of Nueva Cadiz beads from Tiahuanaco: bluish, dark blue, blackish, and greenish (nos. 26, 28, 29, 31) (photo: Saraí Barreiro Argüelles).

PROVENIENCE ANALYSIS

In our approach to the origin of Nueva Cadiz and associated beads, we focused on elements that researchers have used as "tracers" to infer glass provenience. While flux compositions show some regional variations, trace elements in sand are most useful for differentiating glassmaking regions or centers. Among the most eloquent tracers are aluminum found in kaolinite and feldspar, zirconium and hafnium that co-occur in zircon, titanium in rutile, and the cortege of rare earth elements[1] (REE) that concentrate in monazite (Cagno et al. 2012; Coutinho et al. 2021; Degryse and Shortland 2020; De Raedt et al. 2001; Freestone 2005; Koleini et al. 2019; Wedepohl and Simon 2010).

We defined the "diagnostic range" of these elements in the Venezuela and Tiahuanaco beads, i.e., the concentrations that characterize them. The strictest range includes 28 out of 40 beads, and six other beads show a variant, so that our diagnostic ranges account for 85% of our sample. The Nueva Cadiz range falls at the low end of soda glasses of the 15th-17th centuries with proven provenience, so only its upper boundary required definition.

We then compared our diagnostic range with published data on soda glasses in hollowware of the 15th-17th centuries with proven provenience. De Raedt et al. (2001) and Cagno et al. (2012) distinguished Venetian glasses from those made in Antwerp. Similarly, Cagno et al. (2012) distinguished glasses made in Venice and in Altare, a glass center in Liguria, while Coutinho et al. (2016, 2021) separated glasses made in Portugal and Grenada, an Andalusian production center, from Venetian imports. These references cover several of the proposed origins of Nueva Cadiz beads.

Bead studies also provided comparative data. We established diagnostic ranges for glasses from early 17th-century beadmaking workshops in Rouen, Amsterdam, and London (Dussubieux 2009; Dussubieux and Karklins 2016). Interestingly, three Amsterdam samples correlate with Venetian *cristallo*. We also consulted data on French beads of the 17th-18th centuries found around Lake Michigan (Walder 2015). Table 5 synthesizes these results.

Potash

As we have shown, ratios among flux ingredients identify different European glassmaking traditions (Coutinho et al. 2021; Gratuze and Janssens 2004; Šmit et al. 2004; Wedepohl and Simon 2010). Additionally, researchers have associated high phosphorus (1%-3%) with the use of northern European wood ash, while high strontium (> 1000 ppm) points to kelp ash, and chlorine (> 0.5%) denotes Mediterranean soda ash (Degryse and Shortland 2020; Dungworth 2013; Stern 2017; Verità and Zecchin 2009). In our sample, all indicators are consistent with the use of Mediterranean soda ash.

Within the range of Mediterranean soda-ash flux, potash (K_2O) levels of 1.5%-3.0% have been associated with Levantine soda-plant ash used in Venice, and 4.5%-7.5% with Spanish soda-plant ash or barilla used in western Europe (Cagno et al. 2012). The Venezuela beads fall between these ranges (1.9%-4.4%) and we cannot draw any conclusion. In the Tiahuanaco sample, however, small beads, chevron beads, and five large Nueva Cadiz beads (nos. 22, 23, 24, 28, 29) straddle the upper end of the Levantine range (2.1%-3.2%). In contrast, four large Nueva Cadiz specimens (nos. 25, 27, 30, 31) have potash at Spanish barilla levels (3.9%-6.4%). Another (no. 26) has higher potash (9.8%), and its low strontium and high phosphorus indicate the presence of some wood ash in its flux.

Titanium and Aluminum

In the last ten years, researchers have come to realize that each soda-glass center had preferred sources of sand or gravel that carried distinctive geochemical tracers into glass. Titanium is a tracer in studies of Italian and Iberian glasses (Biron and Verità 2012; Cagno et al. 2012; Coutinho et al. 2016, 2021). In our study sample, the diagnostic range for titanium is 113-521 ppm in beads from Venezuela, and 167-447 ppm in those from Tiahuanaco. This range is low, and excludes all our comparisons except Venice and low-titanium glasses from Rouen.

We explored the overlap with Rouen low-titanium samples, by comparing the ratio of titanium to zirconium (Ti/Zr). This ratio in our sample averages 16.4:1. In Rouen glasses, it averages 5.6:1 in samples with low titanium, and 4.0:1 in those with high titanium. This difference suggests different sand sources for the Rouen and Nueva Cadiz beads.

Table 5. Diagnostic Ranges for Compared Elements in Glasses from Venezuela, Tiahuanaco, and Proven European Proveniences.

	Al_2O_3	Ti (ppm)	Zr (ppm)	Hf (ppm)	Nd (ppm)	K_2O
Venezuela ***(diagnostic range)***	***(0.5%-1.3%)***	***(113-282)***	***(5.8-31.4)***	***(0.18-0.89)***	***(0.99-2.97)***	***(1.9%-4.3%)***
Colorless (nos. 1, 2)	0.5%-0.6%	113	5.8	0.18	0.99-1.03	2.7%
Blue (nos. 3-5)	0.9%-1.0%	197-206	10.8-11.0	0.33-0.34	1.68-1.74	3.0%-3.2%
Green (no. 6)	1.2%	255	14.8	0.46	2.47	2.9%
White/blue/white (no. 7)	0.9%-1.7%	240-521	19.1-31.4	0.57-0.89	2.27-2.97	4.1%-4.4%
Nueva Cadiz (nos. 8-10)	0.8%-1.3%	168-282	10.6-16.4	0.34-0.46	1.61-2.12	1.9%-4.3%
Tiahuanaco ***(diagnostic range)***	***(0.6%-1.3%)***	***(167-447)***	***(8.7-29.5)***	***(0.23-0.85)***	***(1.52-3.46)***	***(2.2%-3.6%)***
Nueva Cadiz 1 (n=7)	0.6%-1.0%	217-331	8.7-16.2	0.23-0.48	1.52-2.30	2.2%-3.4%
Nueva Cadiz 1a (nos. 25-27)						4.0%-9.8%
Nueva Cadiz 2 (no. 22)	1.0%-1.2%	370-452	15.9-17.9	0.39-0.49	2.24-2.80	2.7%-3.3%
Nueva Cadiz 3 (no. 29)	1.4%-1.7%	632-771	23.8-29.5	0.68-0.85	2.64-3.46	4.1%-4.7%
Nueva Cadiz 4 (no. 31)	1.1%-1.3%	424-523	18.6-23.0	0.57-0.70	2.52-3.12	5.3%-7.1%
Small, 3 layers (n=17)	0.9%-1.3%	167-447	9.9-15.9	0.23-0.51	1.71-2.87	2.4%-3.6%
Chevrons (nos. 32, 33)	1.0%-1.6%	308-647	12.3-23.0	0.35-0.63	2.12-3.12	2.0%-4.0%
Round (no. 3)	1.3%	470	13.3	0.48	2.73	2.7%
Small, high-Al_2O_3 (no. 5)	3.0%	871	33.4	0.84	4.30	1.9%
Small, HLLA (nos. 7, 14, 15)	2.9%-4.4%	583-1448	88.5-135.1	2.81-4.11	10.9-12.9	3.1%-4.3%
Glasses with known provenience						
Venice *cristallo*	0.6%-1.1%		~10-18	~0.2-0.3		2.5%-3.2%
Venice *vitrum blanchum*	0.8%-2.1%	< 600	~18-50	~0.4-0.7		1.9%-3.4%
Antwerp *cristallo*	1.4%-1.8%		~10-20	~0.8-2.1		2.5%-3.8%
Antwerp *vitrum blanchum*	1.2%-1.4%		~20-35	~0.25-0.45		1.8%-2.6%
Antwerp *façon de Venise*	1.3%-1.7%		~35-120	~1.2-1.3		4.3%-6.7%
Altare	2.1%-7.8%	~500-1500	~20-170			1.1%-7.5%
Grenada	2.1%-4.2%	~600-1350				5.7%-6.9%
Portugal	1.8%-6.1%	~370-750	225-232	5.8-5.9	7.1-32.3	2.0%-6.9%
Rouen 1 (n=9)	0.7%-2.0%	1147-2170	321-558	9.37-16.67	3.78-15.08	2.9%-4.7%
Rouen 2 (n=4)	1.0%-1.3%	328-383	44-68	3.03-3.89	1.24-1.94	3.0%-7.3%
Amsterdam 1 (n=13)	1.4%-2.9%	247-725	21.7-191.2	0.70-5.45	3.49-7.95	2.3%-6.4%
Amsterdam 2 (n=3) (cf. Venice)	1.1%-1.5%	124-189	9.6-12.1	0.33-0.42	1.76-2.19	2.7%-3.2%
London	1.2%-2.6%	248-943	55-139	1.39-4.34	3.94-18.06	3.2%-5.6%
French beads	~1.0%-3.0%	~28-1288	~11-48	≤ 2	~2.6	~2.0%-8.2%

Dilution effect due to the addition of colorants was corrected by dividing the concentrations by (SiO_2 + Na_2O + MgO + Al_2O_3 + K_2O + CaO + MnO + Fe_2O_3) unless the glass was colorless. Values in gray and amber cells fall outside the Venezuela and Tiahuanaco diagnostic range. *See* endnote 2 for data sources.

Alumina (Al_2O_3) levels in glass follow broad regional patterns. European soda glasses tend to have less than 4%, with regional variations that researchers use in provenience studies (Cagno et al. 2012; Coutinho et al. 2016; Dussubieux, Gratuze, and Blet-Lemarquand 2010; Koleini et al. 2019). At the lower end of the scale, Venetian *cristallo* has less than 1%. Other high-quality Venice and Antwerp glasses, known as *vitrum blanchum* and *façon de Venise*, have 1%-2%. At the upper end of the scale, glasses made in the western Mediterranean – in Altare, Grenada, and Portugal – attain 2%-4% alumina. Our references for Rouen and Amsterdam have wide brackets, due to a comprehensive sampling strategy. Glasses from Rouen show 1%-3% Al_2O_3, while those from Amsterdam contain 2%-5%.

In our sample, the diagnostic range for alumina is 0.5%-1.3%, similar to that of Venetian *cristallo*. It partially overlaps the Antwerp and Rouen ranges of 1%-2%. Alumina levels are significantly higher in beads from Altare, Grenada, Portugal, Amsterdam, London, and France.

Neodymium, Zirconium, and Hafnium

Neodymium is a rare earth element (REE) whose concentration in glass is broadly proportional to REE levels in general. Its diagnostic range in Nueva Cadiz and associated beads (1.0-3.5 ppm) is significantly lower than available comparisons from Portugal, Rouen, Amsterdam, London, and unsourced French beads. We do not have a comparative value for Venetian glass, but the Nueva Cadiz range overlaps with three Amsterdam samples whose profile is otherwise consistent with Venetian *cristallo*.

Zirconium and hafnium are related elements that occur regionally in similar ratios, but in different concentrations. In the 34 beads that underpin the diagnostic range for Nueva Cadiz and associated beads, we see 6-31 ppm of zirconium and 0.2-0.9 ppm of hafnium. Among our comparative glasses, only Venetian *cristallo* matches these levels, as well as the Amsterdam subgroup resembling Venetian *cristallo*.

Ratios of zirconium to hafnium are also specific to regional sand sources. Nueva Cadiz and associated beads cluster around 34:1, while one outlier, a high-aluminum blue bead from Tiahuanaco, has a ratio of 40:1 (no. 5). This outlier also has very elevated titanium, and we may assign it to a distinct sand source.

HLLA Provenience

The three monochrome blue beads from Tiahuanaco containing HLLA glass show a different sand profile. Levels of zirconium and hafnium are 6-8 times higher in HLLA beads than in the soda-lime glasses. In general, levels of 26 trace elements in HLLA beads are 3-10 times higher than in other samples in our study.[3] High alumina (4.2%-6.8%) in these beads is typical of the western Mediterranean, reported in Altare (3%-5%), Grenada, and southern Portugal (2.6%-4%) (Cagno et al. 2012; Coutinho et al. 2021; Medici et al. 2015). We note that moderate alumina (3.0%-3.9%) also occurs in HLLA windowpane from northern Europe (Schalm et al. 2007). High phosphorus in HLLA beads indicates the use of wood ash as flux, a practice typical of northern Europe. All these indicators point to a separate provenience, but we need more research to identify the origin of these beads.

Provenience Summary

The elements of Nueva Cadiz and associated beads that we compared have diagnostic ranges at the lower end of their European spectrums. Potash levels in most Tiahuanaco beads fit the profile of Levantine soda used in Venice, and the exceptions indicate the use of Spanish soda. Potash levels in Venezuela beads, however, fall between the Levantine and Spanish ranges.

Titanium and alumina comparisons preclude a western Mediterranean origin for Nueva Cadiz and associated beads. While Antwerp alumina correlates with our beads, we lack data on titanium to confirm this. Venice stands out as the best match.

The zirconium and hafnium levels only match Venetian *cristallo* and the three Amsterdam samples whose profile is consistent with Venetian *cristallo*. Neodymium also matches the three Amsterdam samples, but we lack comparative data on this element for Venice, Antwerp, and several other glass centers.

Available data thus favor Venice as the best match for Nueva Cadiz and associated beads, but we emphasize the need for deeper analysis to verify our comparisons. We also emphasize the need for fuller data from Venice and Antwerp, and possibly from Paris that is missing from our list of comparative references.

CONCLUSION

Nueva Cadiz beads have fascinated researchers for their early arrival in the Americas, their disappearance about 1560-1585, their sophisticated technology, and their unresolved provenience. We have studied the chemistry of two collections, one with a solid archaeological context and the other taken from a site with little regard for its documentation. Both collections emanate from the same beadmaking tradition. While the Venezuela sample provides an early view of Nueva Cadiz and associated beads (ca. 1500-1540), the Tiahuanaco sample shows its later 16th-century development.

Contrary to the perception that colored glasses did not require high-quality sand, these beads were made using sand with a 97%-98.5% silica content. Such purity is typical of many glass beads, which casts beadmaking as a branch of the refined soda-glass industry that arose in Italy and spread throughout Europe in the 15th-17th centuries. Despite their reliance on soda glassmakers, Nueva Cadiz beadmakers controlled many steps of the manufacturing process. They divided each glass batch into three lots to color them turquoise, dark blue, and white, and assembled the colored glasses into production tubes before starting a new glass batch. They did not practice an economy of scale by coloring an entire glass batch the same color, which would have resulted in a different glass batch for each color of a bead. They made the most of their raw materials, as shown by surplus turquoise stock converted to dark blue. A similar workplace organization underlies both collections.

The beads shed light on the use of tin and lead as an opacifying agent. By preventing tin from dissolving in molten glass, lead favors the formation of tin crystals that perform the opacifying role. Beadmakers adjusted opacifier doses in different bead layers to create a mirror-like effect, allowing light to reflect off the white middle layer. They used tin and lead at 25% concentration to create the reflecting white middle layer, at 7.5% to opacify the dark blue inner layer, and at 0.6%-2.2% to create shimmering in the outer layer. The prismatic planes of the square-sectioned bead diffract light and enhance its shimmering effect.

Beadmakers used cobalt from several mines to create the dark blue color in the Venezuela beads, but only cobalt from Schneeberg for the Tiahuanaco beads. Together with the Schneeberg cobalt monopoly, the presence of HLLA glass shows a northern European influence in the Tiahuanaco sample. The HLLA beads reveal a previously unknown 16th-century beadmaking tradition, characterized by high-alumina sand, high-phosphorus flux, cobalt colorant from unidentified mines, and a peculiar shape with bulging sides. Despite their northern European influences, HLLA beads entered the same transatlantic networks as Nueva Cadiz beads.

Regarding the provenience of Nueva Cadiz and associated beads, the flux in Venezuela beads falls between Levantine and Spanish diagnostic ranges. Most Tiahuanaco beads contain Levantine soda-plant ash but some have Spanish barilla. In Europe, only Venice had access to Levantine soda ash. As for sand-related elements, levels of alumina, titanium, zirconium, hafnium, and neodymium exclude a western Mediterranean origin, and cast doubt on Amsterdam, London, Rouen, and other French bead origins. In Europe, Venice stood out for its selective use of crushed river cobbles as a silica source. In the absence of full comparative data, however, we cannot exclude Antwerp or Paris as possible origins. Based on available data, Venice stands as the best candidate as the source of Nueva Cadiz and associated beads, but we emphasize the need for more analyses.

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ENDNOTES

1. The rare earth elements, mostly found in the lanthanoid group at the bottom of the periodic table, are Y, Sc, La, Ce, Pr, Nd, Pm, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu.

2. Venice, Antwerp: De Raedt et al. 2001; Venice *vitrum blanchum* titanium: Biron and Verità 2012; Altare: Cagno et al. 2012; Grenada: Coutinho et al. 2021; Portugal: Coutinho et al. 2016; Rouen: Dussubieux 2009, data for 13 of 28 glasses; Amsterdam, London: Dussubieux and Karklins 2016, data for 16 out of 19 glasses; French beads, Walder 2015.

3. Trace elements, including rare earth elements, occurring at high levels in these three beads are Li, B, Ti, V, Rb, Zr, Cs, Ba, La, Ce, Pr, Ta, Y, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Hf, and Th. Titanium (Ti) deviates somewhat with a high level in bead no. 5 and average in no. 7.

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