

Journal of Archaeological Science 35 (2008) 398-411

Archaeological SCIENCE

http://www.elsevier.com/locate/jas

Degradation processes in colourless Roman glass: cases from the Bocholtz burial

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Received 6 September 2006; received in revised form 28 March 2007; accepted 19 April 2007

Abstract

A group of Roman glass objects from the Bocholtz burial in the SW of Limburg (The Netherlands) was found to have been subject to varying degrees of degradation. Many of the 25 colourless glass objects were fragmented to pieces <0.1 cm ("sugared"), whereas the three transparent blue-green glass objects were in near-pristine state. Analyses using SEM, XRF and EDS revealed that the fragmentation was the direct result of the intense leaching of Na₂O from the glass structure and its replacement with water. The resulting gel layers with low-Na₂O contents and large amounts of water are sensitive to cracking when they dry out. Thin-walled glass appears to be less sensitive to cracking from the resulting stress than thick-walled glass. Local differences in the moisture regime during burial also influence the severity of the glass degradation. Glass with low concentrations of CaO seems to be the most sensitive to this form of degradation. The typical blue-green Roman glass is less sensitive as it generally has considerably higher concentrations of CaO.

"Sugared" glass has – until now – only been reported in a small number of high-status burials. However, glass fragments that are degraded as strongly as some of the objects in the Bocholtz burial may have been overlooked in other archaeological settings (e.g. settlements), especially in coarse-grained soils or soils containing debris or rock fragments. As a result, the archaeological record for Roman glass may be biased towards the "typical" blue-green transparent glass objects.

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Keywords: Gel layer; Glass; Leaching; Degradation; Glass colour; "Sugaring"; Soil moisture

1. Introduction

Roman glass is widely considered to be well-preserved in the archaeological record (Freestone, 2001; Newton and Davidson, 1989). The lack of visible degradation — apart from irisation — is often compared with medieval glass, which can show the results of a range of different degradation processes (Dal Bianco et al., 2005; Geilmann, 1956). This difference is attributed to

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Roman glass being a soda-silica-lime glass, whereas medieval glass usually is a potassium-silica-lime glass. The greater tendency of potassium — compared to sodium — to leach from the glass results in a greater susceptibility of medieval glass to leaching, exfoliation, etc. The soda-silica-lime glass used during Roman times is much less susceptible to leaching and associated degradation features.

Despite this, completely disintegrated glass objects were found at a group of Roman burials in the Netherlands (Esch, excavated between 1952 and 1960, Van den Hurk, 1986; Ypey, 1965). With all fragments of specific objects as small as 0.1 cm or less, the level of fragmentation far exceeded the damage normal for breakage or ground pressure (with at least

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 $^{0305\}text{-}4403/\$$ - see front matter \circledast 2007 Elsevier Ltd. All rights reserved. doi:10.1016/j.jas.2007.04.008

a considerable portion of sherds in the cm-range). Similar cases are known from other locations in the Netherlands (Nijmegen and Maastricht-Belfort: Isings, 1971; Koster, 1997, 2005, Belgium, Haspengouw-Tumuli: Cosyns, 2004; Mariën, 1994; Massart, 2001 and Turkey: Sagalassos, Patrick Degryze, personal communication). This type of degradation seems to be restricted to colourless glass objects, since the well-known green or bluish-green glass does not seem to be affected. As a result of its white, grainy appearance, this type of degraded glass is known among Dutch archaeologists as "sugar glass", and the process as "sugaring".

According to Peyches (1965) and Ypey (1965), the disintegration of the Esch glass objects was caused by leaching of Na⁺. The leaching of Na⁺ from soda-silica-lime glass results in the formation of an outer so-called gel layer, which consists mainly of the remaining glass components (SiO₂ and CaO) and water. The Na⁺ is effectively replaced by H_3O^+ (Newton, 1984; Shelby, 1997). Tests by Peyches (1965) have shown that such gel layers may cause fragmentation of the glass due to shrinkage as it dries out. Several questions remain regarding "sugaring", the foremost being: why only colourless glass seems to be affected and why - within the group of colourless glass objects - some objects have suffered much more damage than others. Some bowls and plates from the Esch burial were for example sugared whereas others had an almost pristine appearance (Ypey, 1965). Also, it remains unclear whether this type of degradation was restricted to certain types of soils: the few reported cases seem to be associated with well-drained soils, but there is no further information available on soil conditions. On a more general level, the question arises as to how common this form of degradation is.

A chance find in 2003 of a late 2nd - early 3rd century Roman burial in Bocholtz, The Netherlands (De Groot, 2006; see Fig. 1), provided an excellent opportunity to study the degradation of Roman glass and to assess the reasons why differences in degradation occur. Apart from a large number of bronze and iron objects, the burial contained 28 glass plates, bottles and jugs; see Fig. 2 for an overview of the objects in the burial chamber. The glass objects showed large differences in form and degree of degradation, ranging from seemingly unaffected to totally disintegrated. Whereas the unaffected glass could be lifted and recovered by hand, the objects most strongly affected by degradation (i.e. the totally disintegrated ones) were excavated by lifting en bloc. The identification of the number and type of glass objects in some of these blocks was only possible using medical CT scans (De Groot, 2006; Jansen et al., 2006). Some of these objects were subsequently excavated (requiring impregnation with paraloid and/or polyurethane), while others were left inside the blocks. Since all objects are from a single site measuring less than 3×3 m, we can assume more or less homogeneous soil conditions for all objects with regard to moisture regime, Eh, grain size and pH. Variation in these parameters was not, however, tested and small-scale variations may occur. Such variations are still expected to be minor compared to variation between sites from different regions, however. Overall, differences in degradation can indicated.

therefore be attributed for the most part to object-related properties like glass composition and shape. Differences in soil composition may be mostly disregarded. Extrapolation to other regions is risky, however. The degradation processes that have been active at the site may be typical for the local soil conditions. Moreover, the presence of other (especially metal) objects near the glass in the burial may have resulted in variations in the soil moisture regime, for example, at the micro-scale.

We analysed the glass from the Bocholtz burial by various means in order to elucidate the processes and causes behind the various types of degradation. First, we made an extensive survey of the macroscopic condition of the glass. Subsequently, we studied the micro-scale effects of degradation on the glass surface and on polished sections using a stereo microscope and a scanning electron microscope (SEM). Third, we performed chemical analyses to determine the glass composition and study the causes and effects of degradation processes. We analysed the bulk chemical composition of each object by X-ray fluorescence spectrometry (XRF) and determined the composition of weathered and unweathered glass in single objects using SEM-mounted energy dispersive spectrometry (EDS) on polished sections.

The main goal of this study is to establish to what extent the degradation process known as "sugaring" is influenced by: (1) the chemical glass composition, (2) morphological object properties, and (3) micro-scale variations in burial conditions.

Fig. 1. Map of the Netherlands showing the location of the Bocholtz excavation. Other sites in the Netherlands where "sugared" glass was found are also





Fig. 2. Drawing of the Bocholtz burial chamber, indicating the position of the various objects (reproduced from De Groot, 2006). The objects inside the sarcophagus are not shown. Numbers refer to the catalogue numbers from De Groot (2006). Nos. 101-128 are glass objects, 201-213 are bronzes, 301-304 iron (though iron nails are not numbered in this drawing), 401 silver and 501 stone objects.

2. Materials and methods

2.1. Site description and history

The Bocholtz burial was discovered in 2003, when a farmer's plough hit the lid of a stone sarcophagus. A rescue excavation by the ROB in November 2003 revealed that the sandstone sarcophagus had been placed in a wood-lined burial chamber of approximately 2.40×2.30 m. At the site, the soil profile consists of a shallow (approximately 60 cm) deep top layer of decalcified loess soil, overlying cretaceous limestone and clay with secondary carbonate encrustations. The burial chamber had been dug through the loess into the top of the limestone/clay layer. A layer of limestone blocks had been placed on top of the wooden roof. Although no traces were found, it is likely that a tumulus had been erected over the burial (De Groot, 2006).

Inside the sarcophagus were the cremated remains of a male who was probably 20- to 34-years-old. A very rich collection of burial gifts was found inside the sarcophagus and in the burial chamber (see Fig. 2). The 17 bronze or composite objects, six iron objects (not counting nails), 28 glass objects and one silver object included several unique pieces. The highstatus of the burial is apparent not only from the large number of high-quality luxury objects, but also because of the absence of pottery. All types of objects that in lower-status burials are made from ceramic are made from metal or glass in the Bocholtz burial. On typological grounds, the burial has been dated to the late 2nd or early 3rd century AD (De Groot, 2006). Fig. 3 shows illustrations of a representative selection of the objects. Some of the objects were in such a poor state that they were not excavated any further. The original shape of these objects could therefore only be determined roughly. Part of the typological classification was based on CT scans of blocks of soil with glass inside (Jansen et al., 2006).

In the first period after the burial, water percolating through the overlying soil washed out the finest fraction (clay) from the profile. This seeped into the burial chamber, partly filling some of the objects and was deposited locally on or beneath the wooden floor. In a later phase — probably after several decades — the wooden structure started to fail, and soil entered the chamber through the sides. Only when most of the objects were buried in a layer of soil material did the roof collapse, depositing the limestone blocks from the roof on top of the



Fig. 3. Drawings of typical glass objects from the Bocholtz burial.

soil material layer that covered and encased most of the objects. As a result, the objects were buried for some 1800 years in decalcified loess that contained some pieces of limestone, directly beneath a layer with blocks of limestone.

2.2. Description of the glass objects

An overview of all glass objects from the Bocholtz burial is given in Table 2, including the catalogue numbers from De Groot (2006). They can be grouped into a few distinct groups. One group - objects101, 102 and 112 - are bottles, one hexagonal and two unguentaria (see drawings in Fig. 3), made from the typical greenish glass found throughout the Roman Empire. Unguentarium 102 was standing inside the sandstone coffin, and is therefore missing from Fig. 2. Two of the bottles (101 and 102) were intact, and could not therefore be sampled for the destructive analyses employed during our study. Unguentarium 112 was shattered by the collapse of the burial chamber, so samples could be taken from this object. The rest of the glass consisted of various types of objects made of colourless or nearly colourless glass. Six were very similar thin-walled cylindrical bottles (nos. 105, 109 and 113-116) with one handle (see Fig. 3). Not all of these bottles were recovered completely, and there is some uncertainty as to whether they were all the same height. Three round-bellied jugs (107, 108 and 110) with trefoil spouts - one (108) showing an additional glass thread decoration – are considered rare in the Netherlands, and in the Rhineland in general (De Groot, 2006), but are quite common in Great Britain (Price and Cottam, 1998). The largest group of glass objects, however, consists of various types of plates and bowls, ranging in diameter from 3.5 to 42.5 cm. Some have solid base rings (e.g. 103, 104; see Fig. 3). Other bowls have no base ring, but could not be assigned to a specific type of bowl because they were sugared and - as a result - too deformed. One plate (106) has a hollow base ring.

2.3. Sampling and sample description

Sherds were selected from each glass object available for sampling. In the case of some of the totally disintegrated objects, this involved identifying the position of the object in the soil block, removing some overlying soil material, and then spooning out the glass fragments. If there were macroscopically visible differences in the degree of fragmentation within a single object, multiple samples were (if possible) taken to represent the various degrees of degradation. Glass standard 7 of the Society for Glass Technology was used as a reference sample for chemical analysis. This sample has a composition close to that of the Bocholtz samples.

The samples were studied macroscopically and with the help of a stereo microscope. The degree of fragmentation and colour of all samples were described. The colour was described in an oblique (perpendicular to the original surface of the object) and transversal (parallel to the original surface of the object) perspective. For a systematic colour description, we used Pantone[®] transparent colour sheets. In the literature,

glass is often described in non-standardized form (with descriptions like "vivid green", "hazy blue-green", etc.). We found that the lack of irreproducible descriptions makes it impossible to compare glass colours from different publications. Most standardized colour sheets (e.g. Munsell[®] colour charts) are made for opaque materials, and are not suitable for describing the colour of transparent materials. We therefore decided to use the Pantone[®] transparent colour sheets.

2.4. Sample treatment

Some of the objects had been strengthened with paraloid or cyanoacrylate glue during excavation from the block of soil. These samples were treated with acetone to remove these components. Some of the samples from the strongly fragmented objects were contaminated with soil material (clay). To remove the clay, the samples were washed with 15 ml 6 N HCl for 15 min and then centrifuged for 10 min at 2800g. Subsequently, the liquid was removed using a Pasteur pipette. If necessary, this procedure was repeated in order to remove as much of the adhering clay as possible. Finally, the samples were rinsed and dried overnight. This method was selected because we deemed it the most effective method of removing clay (thanks to the dispersive effect of acids on clay aggregates), while at the same time preventing too much damage, dissolution or chemical change in the glass composition. Subsamples of the reference sample received the same acetone-HCl treatment to investigate whether this treatment had any effect on the analyses.

2.5. Bulk chemical analyses

The content of volatiles in the glass – the Loss On Ignition at 500 °C (LOI 500) – was determined by weighing the samples before and after they were placed in a 500 °C oven for 5 h.

For the determination of the major element bulk composition by XRF, 0.6 g of sample was mixed with 6 g of a mixture of lithiummetaborate (34%) and lithiumtetraborate (66%). Five-hundred microlitre of 34% lithiumiodide solution in water was added to this mixture, which was then melted at 1200 °C in a Herzog HAG-S to produce a glass bead. The beads are analysed for major and trace elements by X-ray spectroscopy, using an ARL9400 spectrometer with an Rh tube, with full matrix correction The XRF is calibrated using approximately 100 certified geological reference samples. Three reference samples are added to each batch of 50 samples to determine precision (0.5–1% relative standard deviation) and accuracy (1–5% relative standard deviation).

2.6. SEM and SEM–EDS analyses and micromorphology

Subsamples from each sample were used to study crosssections of the glass. The largest fragments of the strongly sugared samples had to be selected, since the powder-like fragments < 0.1 cm were too small to handle for the subsequent processing. The samples were embedded in a mixture of Poly-Pol PS 230 and MEK peroxide. They were wet-polished using sanding sheets of silicon carbide FEPA P#2400 and 4000. Subsequently, they were coated with carbon, and studied using an SEM (JSM 5910LV) with EDS (Vantage, ThermoNoran). To study degradation patterns and gel layer thickness, backscattered electron (BE) images were taken. The greylevel intensities are related to the chemical composition of the specimen, with heavy elements giving a brighter image than lighter elements. Subsequently, three spot measurements were taken with 15–20 kV on the core and three on the leached layer using the EDS system.

In order to study disintegrated glass still present in the soil, part of one of the soil blocks with glass was prepared for micromorphological investigation. This sample contained parts of two objects (small plate or bowl 128 standing on top of large plate 118). This part was strengthened with plaster of Paris and polyurethane foam after separation from the main block. It was then taken to the micromorphological laboratory of Wageningen University, where it was impregnated with synolite. A thin section was made, and studied with a polarization microscope. The remaining impregnated block was polished dry using sanding sheets of 6000, 8000 and 12,000 mesh before analysis by SEM. The block was studied in low vacuum mode (30 Pa) since it was not possible to coat it with carbon.

2.7. Reliability of the chemical analyses

The results of the measurements on the reference sample (Table 1) show that the XRF has good accuracy (absolute differences for Na₂O and SiO₂ <0.9%, for the other elements <0.1). The precision is generally good (std. dev. <0.5%) for most elements, though it is slightly low for SiO₂ (std. dev. approximately 2%). The SEM–EDS system performed remarkably well for semi-quantitative analyses. All parameters show good accuracy with values within one standard deviation of the reported values, with the exception of Al₂O₃ (slightly too low) and Na₂O (too low). This effect can be caused by the impinging electron beam leading to an increase in the temperature of the specimen in the region of the analysis. This

Table 1

Results of the measurements and standard deviations (between brackets) on glass standard 7 of the Society for Glass Technology

	Glass standard 7	XRF results	SEM-EDS results	LOI measurement ^a	
	Reported values	(N = 4)	(N = 6)	(N = 2)	
SiO ₂	72.64 (0.15)	71.82 (2.02)	74.58 (0.74)	n.d.	
TiO ₂	0.042 (0.002)	0.0514 (0.002)	<d.l.< td=""><td>n.d.</td></d.l.<>	n.d.	
Al_2O_3	1.5 (0.07)	1.54 (0.04)	1.21 (0.23)	n.d.	
Fe ₂ O ₃	0.04 (0.002)	0.11 (0.003)	0.16 (0.14)	n.d.	
CaO	11.03 (0.07)	11.04 (0.17)	11.54 (0.27)	n.d.	
MgO	0.14 (0.08)	0.12 (0.01)	0.07 (0.12)	n.d.	
Na ₂ O	13.9 (0.12)	14.34 (0.31)	11.85 (0.49)	n.d.	
K2O	0.43 (0.02)	0.46 (0.03)	0.4 (0.05)	n.d.	
LOI	0.07 (0.03)	n.d.	n.d.	0.21 (-)	

n.d., Not determined; <d.l. is below detection limit.

^a In the reported values, LOI is measured at 550 °C. We heated to 500 °C.

will cause the migration of the alkaline elements (and other volatile elements) away from the region of the glass that is being analysed (Henderson, 1988), thus decreasing the values of the measured alkali concentrations. The precision is also very good (<0.5%) except for SiO₂ (approximately 3% absolute std. dev.). The high precision of the SEM–EDS analyses makes these measurements well-suited to studying variation within our data set, though care should be taken when comparing with other data sets, especially given the lower accuracy for Na₂O.

3. Results

3.1. Macroscopic description

An overview of the glass objects, and the macroscopic description of the samples is presented in Table 2. Only objects 101, 102 and 112 (a hexagonal bottle and two *unguentaria*) have a clear green or blue-green colour, and are therefore

Table 2

Description of all glass objects from the Bocholtz burial

not classified as colourless glass. Objects 104 and 106 (plates) show slight colouring. On these grounds, they may fall in the group of slightly coloured objects as defined by Jackson (2005). Only the clear green and blue-green coloured objects (101, 102 and 112) fall in the class of least fragmented objects (class 1; sherds > 5 cm across). The least fragmented part of plate 104 is also classified in class 1, though the fragmented part falls in class 3 (fragments 0.5–1 cm across). Of the other objects, roughly half fall into class 2 or 3 (fragments 1–5 and 0.5–1 cm across, respectively). The rest fall in class 4 or 5 (fragments 0.5–0.1 or <0.1).

3.2. Description of glass degradation

Strong fragmentation, resulting in disintegration or "sugaring", occurs in a remarkably large number of objects. It is restricted to plates or bowls and to the handles and spouts of the cylindrical bottles. Some of the objects (class 2-3) have broken into fragments which to the naked eye can still be

Object number	Description	Type (from Isings, 1957)	Colour	Colour oblique (Pantone)	Colour transversal (Pantone)	Degree of fragmentation	Remarks	
101	Hexagonal bottle	50	Light green	n.a.	n.a.	1	Not sampled	
102	Unguentarium	82 A2	Light green	n.a.	n.a.	1	Not sampled	
103	Bowl or plate	80	Colourless	406A	413A	4		
104	Bowl or plate; solid part	80	Colourless to slightly green	413A	365A	1	Sample 104.1	
	Bowl or plate; fragmented part			413A	587A	3	Sample 104.2	
105	Cylindrical bottle; body	51b	Colourless	413A	413A	2	Sample 105.1	
	Cylindrical bottle; handle			420A	427A	3	Sample 105.2	
106	Bowl or plate	42b	Colourless to slightly yellow/greenish	587A	365A	2		
107	Jug with trefoil spout	88b	Colourless	413A	365A	2		
108	Jug with trefoil spout	88b	Colourless	400A	365A	2		
109	Cylindrical bottle; body	51b	Colourless	406A	406A	2	Sample 109.1	
	Cylindrical bottle; handle			413A	413A	3	Sample 109.2	
110	Jug with trefoil spout	88b	Colourless	441A	365A	2		
111	Bowl or plate	42b	Colourless	n.a.	n.a.	n.a.	Not sampled	
112	Unguentarium	82 A2	Green-blue	317A	318A	1	_	
113	Cylindrical bottle; body	51b	Colourless	427A	365A	4		
114	Cylindrical bottle; body	51b	Colourless	400A	365A	3		
115	Cylindrical bottle; body	51b	Colourless	406A	365A	2		
116	Cylindrical bottle; body	51b	Colourless	406A	587A	3		
117	Bowl or plate	_	Colourless	413A	587A	3		
118	Bowl or plate	_	Colourless	413A	406A	5		
119	Bowl or plate	_	Colourless	413A	420A	5		
120	Bowl or plate	_	Colourless	403A	400A	5		
121	Bowl or plate	_	Colourless	406A	441A	4		
122	Bowl or plate	_	Colourless	459A	413A	5		
123	Bowl or plate	_	Colourless	413A	427A	4		
124	Bowl or plate	_	Colourless	459A	413A	5		
125	Bowl or plate	_	Colourless	406A	413A	4		
126	Bowl or plate	_	Colourless	413A	441A	4		
127	Bowl or plate	_	n.a.	n.a.	n.a.	n.a.	Not sampled	
128	Bowl or plate	_	Colourless	n.a.	n.a.	n.a.	Not sampled	

The colour and fragmentation are also indicated. n.a., Not available. Degree of fragmentation: 1, no or slight fragmentation (sherds > 5 cm across); 2, moderate fragmentation (sherds 1-5 cm across); 3, strong fragmentation, sherds 0.5-1 cm across; 4, very strong fragmentation, sherds 0.5-0.1 cm across; 5, total disintegration, powder with fragments only sporadically approaching 0.1 cm across. Classes 3-5 are also referred to as "sugar" (or "sugared") glass. Objects 117-128 were "sugared" to such an extent that they were not excavated from the blocks of soil. Their general shape was determined using CT scans, but they could not be classified according to Isings (1957).

recognized as glass, but the worst cases of "sugaring" (class 5) have disintegrated into glass fragments that can hardly be recognized as such (<0.1 cm; Fig. 4a). In the soil blocks, the fragments appear to be still more or less in situ, and together they still more or less delineate the shape of the original object (Fig. 4a–c). However, given the loss of internal cohesion resulting from fracturing, deformation seems to have occurred due to soil movement or compaction. In one case, a bowl (128) seemed to have slumped over its base ring with the base ring protruding through the bottom.

As stated above, plates and bowls are usually disintegrated uniformly. The only exception is plate 104. Most of this plate was barely degraded, but one part – which had been directly beneath bronze jug 203 while buried – was strongly fragmented. The area around the disintegrated part contained some features that may be interpreted as initial or partial disintegration. The most remarkable feature is the occurrence of several short slightly curved isolated fractures (e.g. Fig. 4d).

SEM-analyses of cross-sections through the glass samples show a considerable variation in the thickness of the gel layers that have formed due to the leaching of Na. In SEM backscatter images they have a distinctly darker colour than the unweathered core pictures due to the lack of Na. Both gel layer and core seem homogeneous, with a clear, sharp boundary between them (Fig. 5). In some objects, the gel layer has a uniform thickness, with the boundary between gel layer and core parallel to the surface of the fragments (Fig. 5a). In other objects, the gel layer varies in thickness (Fig. 5b). These fragments suggest that gel layers may be thinner than 10 μ m, but may also reach thicknesses of >500 μ m. Even then, the thickness of the gel layer could have been underestimated, especially in the strongly fragmented objects, as the outer part of the gel layer may have broken away from the sampled fragment.

Gel layers show considerable fracturing. Some of the fractures may have resulted from water loss in the vacuum of the SEM. However, this is clearly not the case with the V-shaped cracks that occur at most steep-angled corners of the glass fragments (Fig. 5a). Samples with thicker gel layers often have multiple fractures, not all of them V-shaped. In several cases, major fractures run from the gel layers through the unweathered core (Fig. 5b). Such fractures seem to occur most frequently in thin glass, especially in samples from the bodies of the cylindrical bottles.

Backscatter images of the impregnated soil block show that the disintegrated objects in the soil consist almost completely of strongly fractured gel layer (Fig. 5c). Remnants of the cores, forming relatively large fragments, occur only sporadically (Fig. 5d).

3.3. Chemistry of weathered and unweathered glass

SEM-EDS analyses of the gel layer and the core show that core Na_2O contents lie in the range 12-18 wt%, whereas these



Fig. 4. Various aspects of disintegrated ("sugar") glass. Scale bars in A, C and D are 1 mm. (A) Disintegrated glass in situ in soil material. (B) Fragmentation pattern in small bowl (object no. 103) during excavation. The glass was impregnated with paraloid to prevent it from disintegrating any further. (C) Thin section showing disintegrated glass (g) of two stacked objects (128 and 118), with soil material and a piece of calcrete (c). (D) Single fracture with both ends in the glass (indicated by arrows); surface of plate 104 close to disintegrated part.



Fig. 5. Backscatter Electron (BE) images of glass in cross-cut samples. (A) Thin gel layer with shrinkage cracks at the edges of the sherd. (B) Thick gel layer with small remaining core. The gel layer is strongly fractured, with a major crack continuing from the gel layer through the core. (C, D) BE images of polished block with glass and soil in situ, showing massive disintegration. Black is pore space, filled with resin. In D, no unweathered core can be seen, C shows some fragments with small pieces of core remaining.

contents have fallen to <3 wt% in the gel layers (Fig. 6; Table 3). Two samples have higher contents (3–6 wt%) in their gel layers: the blue-green *unguentarium* (no. 112) and the non-fragmented part of plate 104. It is puzzling to note that the other sample from plate 104 – from the fragmented part – has Na₂O <3 wt%, like the other objects. Despite this, it is clear that the gel layers have a more or less constant composition. The CaO contents in the gel layer are elevated compared to the core (see below, however).

3.4. Bulk glass composition

The effects of leaching on the bulk composition of the glass – analysed by XRF – are apparent from Fig. 7. Some objects have Na₂O contents close to the values of the core (18–20 wt%), while others show a massive loss of bulk Na₂O. Apparently, these objects consist mostly of gel layer and have hardly any unweathered core left. In the strongly weathered glass, the CaO contents seem to decrease with decreasing Na₂O. At first glance the bulk CaO contents seem at odds with the EDS measurements, as they have decreased considerably in the low-Na₂O-samples, whereas the EDS measurements showed a CaO-increase. The alternative explanation that the strongly weathered glass had lower CaO prior to weathering is not supported by SEM–EDS measurements of the unweathered core. Here, the strongly



Fig. 6. Scatterplot of CaO and Na₂O of the cores and gel layers of each object sampled. The core measurements are indicated with a dark grey, the gel layer measurements with a light grey field. Na₂O contents of the gel layer is decreased dramatically, but it never reaches zero. Broken lines indicate the trend towards lower Na₂O during leaching. Arrows indicate the weathering path of samples with relatively higher Na₂O contents in the gel layer.

leached samples are in the same CaO range as the rest of the samples.

The apparent increase in CaO contents in gel layers as determined by EDS is probably a result of closed-sum effects. The loss of Na_2O results automatically in an increase in the

Table 3	
Results of the chemical analyses	

other elements because the sum of the measured elements is recalculated to 100 wt%. The XRF-measurements are not, however, that strongly affected by the closed-sum effect, as the loss of Na₂O is balanced at least partly by water in the gaps in the crystal structure. This is analysed as LOI 500. A

Sample	XRF (wt%)											
	SiO ₂	Al_2O_3	TiO ₂	CaO	MgO	Na ₂ O	K ₂ O	Fe ₂ O ₃	MnO	P_2O_5	LOI 500 (wt%)	SUM (wt%)
103	69.60	1.74	0.07	5.47	0.28	18.66	0.37	0.31	0.02	0.04	2.31	98.86
104.1	69.96	1.81	0.08	5.60	0.29	18.79	0.38	0.34	0.02	0.05	0.64	97.96
104.2	70.27	1.83	0.07	5.74	0.29	18.82	0.40	0.33	0.01	0.03	1.07	98.86
105.1	68.98	1.80	0.07	5.12	0.30	19.24	0.34	0.34	0.01	0.03	3.16	99.40
105.2	69.94	1.82	0.07	5.15	0.31	20.66	0.36	0.35	0.01	0.04	1.09	99.80
106	66.12	2.16	0.15	5.77	0.57	19.61	0.47	0.59	0.02	0.06	4.17	99.67
107	69.15	2.11	0.09	5.90	0.35	18.96	0.50	0.49	0.12	0.05	1.69	99.40
108	68.18	2.01	0.09	5.71	0.36	19.43	0.48	0.46	0.07	0.04	1.83	98.65
109.1	69.14	1.83	0.08	5.31	0.32	18.76	0.43	0.37	0.01	0.04	3.01	99.29
109.2	71.00	1.89	0.08	5.42	0.34	20.35	0.43	0.37	0.01	0.04	0.77	100.71
110	70.40	1.80	0.08	5.27	0.28	17.13	0.37	0.37	0.01	0.03	3.00	98.74
112	70.11	2.32	0.10	7.11	0.45	18.60	0.58	0.52	0.34	0.10	0.46	100.70
113	71.32	1.99	0.08	5.11	0.33	19.63	0.40	0.32	0.01	0.05	2.53	101.76
114	70.14	1.78	0.07	5.27	0.29	18.13	0.38	0.36	0.01	0.03	3.22	99.69
115	69.71	1.79	0.07	5.33	0.26	19.89	0.41	0.36	0.02	0.02	1.65	99.52
116	71.02	1.89	0.07	5.45	0.32	18.24	0.36	0.32	0.01	0.04	1.67	99.40
117	69.75	1.95	0.08	5.58	0.39	19.83	0.46	0.40	0.01	0.03	1.31	99.79
118	73.73	2.14	0.10	4.43	0.37	4.54	0.16	0.43	0.01	0.03	9.46	95.41
119	73.25	2.91	0.17	3.95	0.42	1.48	0.24	0.58	0.01	0.03	11.08	94.11
120	70.30	2.01	0.09	5.28	0.37	12.33	0.28	0.36	0.02	0.04	7.23	98.31
121	70.82	1.81	0.07	5.50	0.29	18.02	0.39	0.31	0.02	0.04	2.73	100.02
122	71.89	2.04	0.09	4.68	0.39	5.57	0.19	0.42	0.01	0.03	9.52	94.83
123	71.40	1.93	0.08	5.69	0.38	14.82	0.33	0.36	0.02	0.04	6.21	101.24
124	69.10	1.87	0.07	5.62	0.35	16.75	0.34	0.34	0.02	0.05	5.18	99.69
125	71.26	1.80	0.07	5.54	0.32	17.77	0.39	0.31	0.02	0.05	3.07	100.60
126	68.88	1.74	0.06	5.38	0.31	15.83	0.36	0.31	0.02	0.03	6.82	99.77
Sample	SEM-ED	S core (std	dev.; N =	3)								
	Na_2O (wt	%) Al ₂	O ₃ (wt%)	SiO_2 (w	/t%)	K ₂ O (wt%)	CaO (wt	%) N	MgO (wt%)	Fe_2O_3 (wt)	%) MnO (wt%)	Cl (wt%)
103	16.48 (0.1	9) 1.54	4 (0.10)	74.48 (0	0.10)	0.36 (0.07)	5.36 (0.0	04) ().29 (0.03)	<d.l.< td=""><td><d.1.< td=""><td>1.47 (0.04)</td></d.1.<></td></d.l.<>	<d.1.< td=""><td>1.47 (0.04)</td></d.1.<>	1.47 (0.04)
104.1	16.03 (0.1	1) 1.70	0 (0.08)	73.89 (0	0.21)	0.42 (0.09)	5.96 (0.1	(9)).21 (0.08)	0.38 (0.17)	0.11 (0.06)	1.34 (0.07)
104.2	15.37 (0.1	5) 1.30	0 (0.09)	74.93 (0	0.35)	0.34 (0.09)	6.08 (0.2	21) ().14 (0.10)	0.4 (0.13)	0.09 (0.02)	1.38 (0.02)
105.1	18.53 (0.2	6) 1.38	8 (0.02)	72.84 (0).33)	0.31 (0.04)	4.93 (0.1	.0) ().11 (0.12)	0.41 (0.14)	0.09 (0.06)	1.43 (0.05)
106	15.60 (0.1	7) 1.98	8 (0.06)	74.16 (0	0.03)	0.39 (0.08)	5.68 (0.2	24) <	<d.l.< td=""><td>0.62 (0.15)</td><td>0.09 (0.10)</td><td>1.51 (0.05)</td></d.l.<>	0.62 (0.15)	0.09 (0.10)	1.51 (0.05)
107	13.78 (0.2	7) 1.7.	3 (0.02)	75.45 (0).26)	0.52 (0.03)	6.22 (0.1	2) <	<d.l.< td=""><td>0.71 (0.05)</td><td>0.22 (0.18)</td><td>1.36 (0.06)</td></d.l.<>	0.71 (0.05)	0.22 (0.18)	1.36 (0.06)
108	15.06 (0.3	1) 1.5	7 (0.17)	75.13 (0).23)	0.47 (0.04)	5.77 (0.0)7) <	<d.1.< td=""><td>0.52 (0.06)</td><td>0.19 (0.05)</td><td>1.35 (0.09)</td></d.1.<>	0.52 (0.06)	0.19 (0.05)	1.35 (0.09)
109.1	15.76 (0.2	6) 1.4	7 (0.05)	74.95 (0).11)	0.37 (0.09)	5.34 (0.2	21) <	<d.l.< td=""><td>0.46 (0.19)</td><td>0.10 (0.11)</td><td>1.59 (0.04)</td></d.l.<>	0.46 (0.19)	0.10 (0.11)	1.59 (0.04)
109.2	17.30 (0.1	9) 1.39	9 (0.09)	73.71 (0).13)	0.44 (0.01)	5.19 (0.0)9) (0.10 (0.06)	0.3 (0.17)	<d.l.< td=""><td>1.54 (0.11)</td></d.l.<>	1.54 (0.11)
110	16.48 (0.1	5) 1.2	1 (0.13)	74.56 (0).24)	0.33 (0.04)	5.36 (0.1	(2)).10 (0.09)	0.42 (0.16)	<d.l.< td=""><td>1.53 (0.09)</td></d.l.<>	1.53 (0.09)
112	13.68 (0.1	6) 1.99	9 (0.27)	74.65 (0	0.39)	0.47 (0.09)	7.00 (0.3	51) <	<d.l.< td=""><td>0.5 (0.08)</td><td>0.38 (0.02)</td><td>1.32 (0.09)</td></d.l.<>	0.5 (0.08)	0.38 (0.02)	1.32 (0.09)
113	17.87 (0.1	1) 1.20	(0.10)	74.07 (0).21)	0.28 (0.03)	4.68 (0.0)9) <	<d.l.< td=""><td>0.28 (0.18)</td><td><d.l.< td=""><td>1.58 (0.02)</td></d.l.<></td></d.l.<>	0.28 (0.18)	<d.l.< td=""><td>1.58 (0.02)</td></d.l.<>	1.58 (0.02)
114	15.65 (0.0	5) 1.3	/ (0.03)	75.48 (0).19)	0.37 (0.03)	5.28 (0.0)/) <	<d.l.< td=""><td>0.32 (0.17)</td><td><d.l.< td=""><td>1.51 (0.05)</td></d.l.<></td></d.l.<>	0.32 (0.17)	<d.l.< td=""><td>1.51 (0.05)</td></d.l.<>	1.51 (0.05)
115	16.08 (0.2	6) 1.28	8 (0.08)	74.92 (0).10)	0.38 (0.03)	5.27 (0.2	22) <	<d.l.< td=""><td>0.43 (0.1)</td><td><d.l.< td=""><td>1.63 (0.05)</td></d.l.<></td></d.l.<>	0.43 (0.1)	<d.l.< td=""><td>1.63 (0.05)</td></d.l.<>	1.63 (0.05)
116	18.06 (0.7	6) I.30	5 (0.02)	73.22 (0	J.58)	0.37 (0.07)	5.08 (0.0)4) (0.20 (0.07)	0.27 (0.27)	<d.l.< td=""><td>1.50 (0.06)</td></d.l.<>	1.50 (0.06)
117	16.70 (0.0	9) 1.40) (0.06)	73.83 (0	0.27)	0.44 (0.06)	5.54 (0.0)) ().26 (0.17)	0.4 (0.05)	<d.l.< td=""><td>1.42 (0.01)</td></d.l.<>	1.42 (0.01)
118	18.14 (0.0	(7) 1.24	4 (0.08)	73.04 (0	J.29)	0.35 (0.04)	5.06 (0.1	(0) ().16 (0.17)	0.45 (0.03)	<d.l.< td=""><td>1.58 (0.02)</td></d.l.<>	1.58 (0.02)
119	17.39 (0.1	9) 1.32	2 (0.08)	73.80 (0).20)	0.34 (0.04)	5.12 (0.1	.9) <	<d.l.< td=""><td>0.44 (0.29)</td><td><d.l.< td=""><td>1.59 (0.11)</td></d.l.<></td></d.l.<>	0.44 (0.29)	<d.l.< td=""><td>1.59 (0.11)</td></d.l.<>	1.59 (0.11)
120	16.61 (0.2	4) 1.5	1 (0.05)	73.95 (0	0.01)	0.38 (0.02)	5.43 (0.0)4) <	<d.l.< td=""><td>0.42 (0.24)</td><td>0.06 (0.01)</td><td>1.67 (0.06)</td></d.l.<>	0.42 (0.24)	0.06 (0.01)	1.67 (0.06)
121	15.21 (0.1	6) I.I() (0.06)	75.54 (0	0.20)	0.36 (0.03)	5.73 (0.0)/) <	<d.l.< td=""><td>0.32 (0.03)</td><td><d.l.< td=""><td>1.69 (0.08)</td></d.l.<></td></d.l.<>	0.32 (0.03)	<d.l.< td=""><td>1.69 (0.08)</td></d.l.<>	1.69 (0.08)
122	18.53 (0.1	a) 1.4.	5 (U.U/)	72.28 (0	J.14)	0.34 (0.01)	5.48 (0.0)4) <	< u.l.	0.3 (0.13)	0.13 (0.004)	1.54 (0.10)
123	10.75 (0.1	δ) 1.3.	5 (0.15) 7 (0.12)	/4.17 ((J.12)	0.36 (0.05)	5.28 (0.1	10) <	< u.l.	0.39 (0.17)	< a.l.	1.09 (0.06)
124	17.04 (0.0	0) 1.4	(0.12)	/3./0 ((J.31)	0.38 (0.05)	5.42 (0.2	(9) <	<u.i.< td=""><td>0.44 (0.19)</td><td>0.14 (0.11)</td><td>1.70 (0.06)</td></u.i.<>	0.44 (0.19)	0.14 (0.11)	1.70 (0.06)
125	16.05 (0.1	(1) 1.19	2 (0.00)	74.05 (0	J.18) J.18)	0.32(0.04)	5.38 (0.1	1) ())	7.09 (0.00) 24.1	0.38 (0.2)	< 0.1.	1.51 (0.05)
120	10.05 (0.1	0) 1.3.	5 (0.17)	/4.00 ((5.10)	0.37 (0.07)	5.41 (0.1	(0)	_u. 1.	0.55 (0.13)	0.03 (0.02)	1.36 (0.11)

Table 3 (continued)

Sample	SEM-EDS gel layer (std. dev.; $N = 3$)									
	Na2O (wt%)	Al ₂ O ₃ (wt%)	SiO ₂ (wt%)	K ₂ O (wt%)	CaO (wt%)	MgO (wt%)	Fe ₂ O ₃ (wt%)	MnO (wt%)	Cl (wt%)	
103	2.12 (2.03)	1.74 (0.10)	86.58 (2.19)	0.34 (0.14)	6.46 (0.10)	0.35 (0.06)	0.57 (0.19)	0.3 (0.06)	1.65 (0.08)	
104.1	4.80 (4.13)	1.81 (0.04)	84.06 (3.89)	0.39 (0.20)	6.61 (0.16)	0.32 (0.12)	0.41 (0.18)	<d.1.< td=""><td>1.56 (0.04)</td></d.1.<>	1.56 (0.04)	
104.2	1.12 (0.84)	1.72 (0.27)	87.57 (0.71)	0.31 (0.04)	6.79 (0.14)	0.33 (0.12)	0.52 (0.17)	0.06 (0.01)	1.60 (0.02)	
105.1	1.92 (1.61)	1.70 (0.14)	87.94 (1.32)	0.17 (0.07)	5.67 (0.07)	0.38 (0.07)	0.34 (0.10)	0.20 (0.03)	1.76 (0.04)	
106	0.90 (0.66)	2.45 (0.39)	86.50 (0.30)	0.26 (0.06)	6.57 (0.11)	0.66 (0.12)	0.80 (0.03)	0.09 (0.04)	1.79 (0.02)	
107	0.46 (0.34)	2.34 (0.26)	87.16 (0.08)	0.24 (0.20)	7.23 (0.10)	0.43 (0.05)	0.58 (0.14)	0.21 (0.08)	1.52 (0.05)	
108	0.24 (0.16)	1.95 (0.03)	88.08 (0.30)	0.20 (0.04)	6.76 (0.23)	0.43 (0.04)	0.57 (0.11)	0.09 (0.04)	1.71 (0.03)	
109.1	1.03 (0.39)	1.48 (0.12)	88.75 (0.50)	0.23 (0.16)	6.18 (0.02)	0.32 (0.22)	0.38 (0.05)	<d.1.< td=""><td>1.72 (0.11)</td></d.1.<>	1.72 (0.11)	
109.2	0.86 (0.56)	1.72 (0.21)	88.37 (0.77)	0.24 (0.04)	6.18 (0.22)	0.29 (0.03)	0.37 (0.25)	0.09 (0.07)	1.88 (0.05)	
110	1.16 (0.18)	1.41 (0.06)	88.37 (0.36)	0.14 (0.05)	6.33 (0.07)	0.19 (0.05)	0.40 (0.09)	0.19 (0.12)	1.86 (0.04)	
112	4.75 (4.87)	2.04 (0.22)	81.47 (5.07)	0.71 (0.03)	8.25 (0.13)	<d.1.< td=""><td>0.56 (0.18)</td><td>0.70 (0.31)</td><td>1.47 (0.15)</td></d.1.<>	0.56 (0.18)	0.70 (0.31)	1.47 (0.15)	
113	0.92 (0.48)	1.67 (0.13)	89.51 (0.24)	0.05 (0.05)	5.52 (0.05)	0.29 (0.06)	0.19 (0.13)	0.01 (0.01)	1.86 (0.05)	
114	1.05 (0.21)	1.61 (0.10)	88.27 (0.27)	0.22 (0.07)	6.17 (0.05)	0.35 (0.11)	0.47 (0.15)	0.10 (0.14)	1.80 (0.11)	
115	1.19 (0.29)	1.66 (0.13)	88.27 (0.66)	0.18 (0.13)	6.00 (0.11)	0.37 (0.06)	0.43 (0.10)	0.05 (0.03)	1.87 (0.02)	
116	1.60 (0.40)	1.92 (0.13)	87.43 (0.32)	0.16 (0.04)	6.14 (0.11)	0.32 (0.10)	0.55 (0.16)	<d.1.< td=""><td>1.86 (0.04)</td></d.1.<>	1.86 (0.04)	
117	1.15 (0.18)	1.88 (0.24)	87.79 (0.21)	0.22 (0.07)	6.37 (0.18)	0.45 (0.04)	0.52 (0.15)	0.08 (0.02)	1.56 (0.04)	
118	1.64 (0.59)	1.43 (0.13)	88.15 (0.61)	0.20 (0.03)	6.03 (0.29)	0.28 (0.06)	0.36 (0.21)	0.12 (0.11)	1.83 (0.06)	
119	1.21 (0.60)	1.72 (0.22)	88.34 (0.90)	0.11 (0.09)	6.04 (0.10)	0.26 (0.06)	0.39 (0.14)	0.07 (0.08)	1.88 (0.09)	
120	1.00 (0.20)	1.88 (0.11)	87.69 (0.15)	0.16 (0.05)	6.40 (0.27)	0.29 (0.07)	0.56 (0.12)	<d.1.< td=""><td>1.98 (0.10)</td></d.1.<>	1.98 (0.10)	
121	0.71 (0.14)	1.55 (0.16)	88.40 (0.26)	0.18 (0.12)	6.41 (0.21)	0.43 (0.04)	0.45 (0.07)	0.11 (0.15)	1.79 (0.06)	
122	1.24 (0.35)	1.81 (0.33)	88.24 (0.13)	0.14 (0.04)	6.15 (0.13)	0.27 (0.11)	0.21 (0.09)	0.13 (0.17)	1.82 (0.04)	
123	0.62 (0.46)	1.62 (0.06)	88.82 (0.57)	0.09 (0.05)	6.04 (0.12)	0.36 (0.13)	0.39 (0.13)	0.15 (0.05)	1.90 (0.06)	
124	1.84 (0.94)	1.57 (0.22)	87.12 (0.80)	0.17 (0.06)	6.49 (0.08)	0.21 (0.11)	0.63 (0.30)	0.20 (0.19)	1.85 (0.10)	
125	0.92 (0.08)	1.41 (0.15)	88.94 (0.39)	0.16 (0.07)	6.16 (0.05)	0.30 (0.11)	0.26 (0.12)	0.09 (0.06)	1.79 (0.09)	
126	1.16 (0.03)	1.73 (0.04)	88.28 (0.09)	0.08 (0.09)	6.26 (0.02)	0.41 (0.11)	0.18 (0.09)	0.11 (0.07)	1.78 (0.12)	

Two subsamples were taken from objects 104, 105 and 109 (see Table 2). The SEM-EDS measurements are based on the average of three spot measurements. <d.1. Indicates that one or more of the three replicate measurements were below the detection limit.

plot of Na₂O versus LOI (Fig. 8) clearly demonstrates that decreased Na₂O results in increased LOI. Moreover, Fig. 9 demonstrates that an increase in bulk SiO₂ in strongly weathered glass correlates with a decrease in CaO. The apparent increase in CaO in Fig. 8 is therefore an artefact caused by recalculating the EDS measurements to 100 wt%.

4. Discussion

4.1. Glass 'sugaring' and the influence of glass composition and object shape

It is remarkable to note that the degree of fragmentation is mostly homogeneous within one object (apart from the exceptions discussed below) and that objects differing in their degree of degradation were found directly next to each other in the soil. This indicates that object-related properties largely determine the degree of fragmentation of the Bocholtz glass objects, while variation in the burial conditions has limited effects. If soil conditions had been the major factor controlling the degree of degradation, it is unlikely that such clear separation would have been seen. One would therefore expect much larger differences in the degree of degradation within individual objects.

The SEM observations demonstrate that the basic process behind the disintegration of glass objects is the leaching of Na₂O. If leaching has been intense, the glass will consist largely of gel layer. In such extreme cases, the glass has lost 12-16 of the original approximately 18 wt% of Na₂O. Moreover, K₂O (not shown) and some fraction of the CaO will also have been lost. This mass loss is partly compensated by incorporation of water in the leached glass structure. The resulting water-rich gel layer is very sensitive to fracturing when the water is lost



Fig. 7. Scatterplot of the bulk contents (XRF-measurements) of CaO and Na_2O . The dotted line surrounds the concentrations of the colourless glass from Jackson (2005) and Silvestri et al. (2005b). Symbols as in Fig. 6.

LOI 500°C (Wt %) Fig. 8. Scatterplot of bulk Na₂O and Loss on Ignition at 500 °C (LOI 500). The loss of sodium from the glass clearly correlates with an increase in water content. Symbols as in Fig. 6.

from the structure due to drying. In a well-drained soil, like the one in Bocholtz, moisture conditions vary on short time-scales, depending on the weather and the seasons. Wet conditions after rainy periods or heavy showers alternate with dry conditions during periods with little rain and strong evapotranspiration.

Fig. 9. Scatterplot of the bulk concentrations of SiO₂ and CaO. The three samples on the right have high SiO₂ due to weathering. The low-CaO contents demonstrate that some CaO is lost relative to SiO₂ during weathering. Symbols as in Fig. 6.

The buried archaeological objects in the soil therefore continuously experience drying and wetting cycles. During such cycles the gel layer will shrink (when dry) and expand (when wet). During shrinkage, stresses develop which concentrate in the interface between gel layer and unaffected glass. Shelby (1997) describes how such stresses can result in the leached layer peeling off the glass. In our case, the gel layer apparently fractures during drying, but instead of peeling off, the fractures run into the unaffected glass, initiating the first stage of fragmentation. In glass manufacture, a great deal of effort goes into preventing stress in the finished glass product. Such stress would result in breakage during manufacture or shortly afterwards. To prevent stress, glass objects are annealed directly after shaping. They are reheated in a special oven, where the temperature is below melting point but still high enough to allow viscous flow and stress relaxation (Newton and Davidson, 1989). It is highly unlikely that the Bocholtz objects (and other "sugared" glass objects) are insufficiently annealed, as they would not have survived manufacture and use, especially not in such great number. Apparently, extensive leaching and gel layer formation in combination with repeated drying and wetting resulted in stresses great enough to cause fragmentation of the objects. Fracturing and gel layer formation may even have turned into a feedback loop, with fracturing causing increased exposure to soil moisture, facilitating increased leaching and the formation of gel layers which fracture more easily, etc. This feedback is corroborated by the correlation between the gel layer thickness - and thus increased leaching of Na₂O - and the level of fragmentation.

The question remains why some objects have undergone much more intensive leaching than others, resulting in much more fragmented objects. A plot of the degree of degradation versus the CaO content in the glass core shows that, in the case of the plates and bowls, stronger disintegration correlates with lower CaO contents (Fig. 10). This confirms that glass with lower CaO contents is more susceptible to leaching of Na₂O as stated by Newton (1984) and Shelby (1997), resulting in gel layer formation and fracturing. This also explains the good quality (and lack of sugaring) of the typical blue-green glass, as this type of glass has systematically higher CaO contents (e.g. 112 as a high CaO outlier in Fig. 9).

However, bottles do not show this correlation and in general show less fragmentation than plates and bowls with the same CaO contents. This suggests that the susceptibility to leaching of Na₂O depends not only on the CaO content, but also on other properties of the glass objects, more specifically on the shape. This is difficult to explain on the basis of purely process - chemical processes. One clue might lie in the degradation pattern in the cylindrical bottles. The spouts and handles of these objects are usually more fragmented than the bodies, even though there are no clear differences in chemical composition. This difference in degree of fragmentation of the cylindrical bottles might be caused by a similar object property-related difference in susceptibility to Na₂O leaching and gel layer formation. The most obvious difference between these bottle parts is the glass thickness, with the thicker parts of the bottles (i.e. handles and spouts) more "sugared" than







Fig. 10. Scatterplot of the degree of fragmentation plotted versus the CaO content of the core. The dotted line separates plates, dishes and bowls from bottles and jugs. The former show a correlation between CaO content and degree of fragmentation. This correlation is not present in the bottles and jugs. Symbols as in Fig. 6. Degree of fragmentation classes as in Table 2.

the thin body. Similarly, bowls and plates are generally much thicker than the other objects and also tend to be more fragmented. This suggests that thicker glass has greater sensitivity to fragmentation.

As a possible explanation, we would advance the suggestion that thin-walled glass objects might be more capable of accommodating pressure differences that occur due to the shrinking of the initial gel layer (e.g. by deforming slightly); thick-walled objects would fracture more easily because they are harder to deform.

4.2. Influence of the burial environment

The burial conditions in which all objects studied here were found can be regarded as typical of temperate, finegrained, well-drained slightly basic soils. Almost all variation in degree of degradation between and within objects can be attributed to differences in glass composition and object shape. The only case where variations in burial conditions may have played a role in causing differences in the degree of degradation is in the disintegration of the part of plate 104 that was beneath a copper/bronze jug, leaving the rest of the plate intact. Possibly, a difference in soil moisture regime beneath the jug caused more severe leaching, resulting in a thicker, more Na₂O-depleted gel layer. This demonstrates that small variations in the burial environment - and more specifically, in the soil moisture regime can have major effects on the degradation of these types of glass object.

Other finds of "sugar glass" from the Netherlands (Esch and Nijmegen) are also from well-drained soils, but from regions which generally have more acidic and coarser-grained soils. It is difficult to predict how glass objects with similar shape and composition as the ones studied here would fare under different soil conditions. In waterlogged conditions, leaching and gel layer formation might progress at a faster rate because of the efficient transport of the dissolution products. However, since no drying-out phases occur, the chances of it resulting in disintegration could be smaller.

4.3. Implications for the archaeological record

The Bocholtz glass composition is not exceptional. The original CaO contents of most objects are low, but still fall in the range of Roman colourless glass from other sites both in the UK and in Italy (Jackson, 2005; Silvestri et al., 2005a) – whereas the contents of Na₂O, SiO₂ and other major elements are perfectly normal for Roman glass in general. It is therefore surprising that the disintegration (or "sugaring") of Roman colourless glass has received so little attention in the literature on glass and - more specifically - on the degradation of glass. Most authors simply state that Roman soda-silica-lime glass has no or very little susceptibility to degradation. A few publications describe the disintegration of the Roman glass from Esch, or include a reference to it in their general description of degradation processes (Newton, 1984; Newton and Davidson, 1989; Peyches, 1965; Shelby, 1997; Van den Hurk, 1986; Ypey, 1965). In addition, some publications describing glass typology or excavations mention sugaring (Isings, 1971; Koster, 1997, 2005; Mariën, 1994; Massart, 2001).

As stated above, the only documented cases of strongly disintegrated ("sugared") glass are from the Netherlands and Belgium. Given the fact that the composition of the Bocholtz glass – and the soil conditions – are not rare, one would expect "sugared" glass to be more common. However, this does not seem to be the case. Hardly any documentation is available either on this type of degradation, or on objects that are degraded in this way. A major reason for this may be that such glass is often not recognized as such during excavations, and therefore not recovered. In fact, the conditions for finding and recovering such strongly disintegrated glass were exceptionally good at the Bocholtz site for several reasons that do not apply to other sites:

- (1) In a burial, whole and complete objects are buried. In most other archaeological contexts, glass will be broken and fragmented. Such fragments are much harder to recognize as archaeological objects.
- (2) Burials are excavated using different methods than settlements, for example, with much more care and attention focused on a relatively small area. This makes it easier to recognize strongly fragmented glass.
- (3) The grain size of the soil material was small, smaller still than most of the glass fragments. Loess consists largely of silt-sized grains (i.e. in the size range $4-63 \mu m$). These

are a lot smaller even than most of the fragments of disintegrated glass, which makes it easier to recognize the glass. In coarser soils, especially those containing large sand grains, gravel, rock fragments or debris, the glass fragments would be difficult to distinguish. From that perspective we were lucky that the burial chamber had been filled in with loess, albeit only partly, before the collapse of the roof with limestone blocks. If the disintegrated glass had been intermingled with the limestone blocks, most of it would probably have gone unrecognized during the excavation.

Based on the above discussion, it is possible that "sugared" glass is in fact relatively common in Roman glass-containing sites, but that it is often not recognized and therefore not recovered. This implies that the archaeological record for Roman glass is biased towards high CaO glasses. Since the typically blue-green glass objects generally have higher CaO contents (Jackson, 2005), their chance of survival in the archaeological record is better. Within the group of low-CaO – i.e. colourless – glass, the bias is towards thin-walled objects, especially bottles, which have better survival chances. Thick glass objects, like several types of plates and bowls, have a much lower chance of survival because they become "sugared". Such objects may therefore have been much more common than is assumed based on their relative numbers.

5. Conclusions

The glass from the Bocholtz burial has deteriorated in several ways, depending on the composition and the shape of the object. Objects with high CaO contents, i.e. the typical bluegreen Roman glass, have thin gel layers. Colourless objects generally have thicker gel layers. Within this group, the CaO content and the shape of the object determine the degree of degradation. The plates and bowls with relatively low-CaO contents are completely disintegrated ("sugared"), whereas the ones with high CaO contents are not strongly fragmented. The bottles, however, are generally less fragmented than plates with similar CaO contents. Localized variations in the soil moisture regime play an additional role in the degree of degradation.

The total disintegration of glass ("sugaring") as described in this paper is virtually absent from the literature on both Roman glass and on degradation of glass in general. Since low-CaO, colourless soda-silica-lime glass occurred throughout the Roman Empire, and since soil conditions like the ones at the Bocholtz site are common, it is likely that "sugaring" actually occurs quite often. The reason for its scarcity in the literature is probably that the strongly degraded glass is often not recognized as such, and is therefore not recovered during excavations.

Acknowledgements

We would like to thank the following people for making this study possible. From the ROB: Tessa de Groot for allowing us to sample the Bocholtz glass objects and for casting a critical eye over the manuscript; and Jaap Kottman for his help in sampling the glass, especially the objects lifted en bloc. The drawings in Fig. 3 were made by Mikko Kriek, based on the work by Jaap Kottman. Thanks also to Rob van Galen and Erik van Vilsteren (both TNO-Built Environment and Geosciences/National Geological Survey) for their work in the lab on the destruction and analysis of the glass samples, and to Luc Megens (ICN) for assistance in polishing the glass cross-sections. The thin section was prepared by Toine Jongmans and Adri van den Berg at the Laboratory for Soil Science and Geology, Wageningen University. We are grateful to Caroline Jackson (University of Sheffield) and Alberta Silvestri (Università de Padova) for providing digital versions of their published compositional data on colourless glass. Thanks to Sue McDonnell for checking the English. Thanks also to three anonymous referees for their comments on a previous draft of the manuscript.

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