

## Archaeological and historical glasses: A bibliometric study

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Glass is one of the materials more widely developed throughout History. In the last decades, it has been stated a growing demand in the application of chemical-physical techniques to obtain more detailed information on technology and production of glasses in past societies. This research field lies within the domain of archaeometry. Results of a bibliometric study undertaken on 201 scientific articles published on ancient and historical glasses between 1987 and 2008 are presented in this paper. The study was carried out with the aim to address the evolution of glass archaeometric investigations in the last 20 years. Date of publication, journal and article types, topic, glass typology, analytical techniques, origin country of authors, and geographic location of samples were analyzed in this study, among other parameters. Resulting data indicate that archaeometric research on glasses has experienced an exponential growth in the period 2000-2008. Roman and Medieval glasses have been the materials more frequently investigated.

*Key words: Glass, archaeometry, bibliometric study.*

### Estudio bibliométrico sobre vidrio arqueológico e histórico

El vidrio es uno de los materiales que más se ha utilizado a lo largo de la historia. En las últimas décadas se ha producido un aumento en la aplicación de técnicas químico-físicas para estudiar de forma más detallada la tecnología y producción de vidrio en las sociedades del pasado. Este tipo de investigación se encuadra en la disciplina conocida como Arqueometría. En este trabajo se ha realizado un estudio bibliométrico que abarca 201 artículos científicos sobre vidrio arqueológico e histórico publicados entre 1987 y 2008. El estudio se llevó a cabo con el objetivo de conocer la evolución de las investigaciones arqueométricas sobre vidrio en los últimos 21 años. Los parámetros analizados en este estudio bibliométrico fueron: fecha de publicación, tipo de revista y de artículo, tema, tipología del vidrio, técnicas analíticas, origen de los autores y localización geográfica de las muestras. Los datos obtenidos indicaron que la investigación arqueométrica sobre vidrio ha experimentado un crecimiento exponencial en el período 2000-2008. Los vidrios romanos y medievales han sido los materiales estudiados más frecuentemente.

*Palabras clave: Vidrio, arqueometría, estudio bibliométrico.*

### 1. INTRODUCTION

Glass is a widely used material due to its singular characteristics. In ancient Egypt, for instance, it was considered more valuable than precious stones. Glass was also very popular in Medieval cathedrals in which stained glass windows occupied large extensions in their walls. Throughout the last decades a growing demand in the application of advanced chemical-physical techniques has been stated in the study of ancient and historical glasses. This research field lies within the more general domain of the so-called Archaeometry, the discipline which engages experimental and human sciences in the study of contextualized ancient objects.

With the aim to know the evolution of glass investigations undertaken under this archaeometric perspective in the last 20 years, a bibliometric study on 201 scientific articles published on ancient and historical glasses has been carried out. The results of such a study are presented in this paper. Either experimental scientists or historians and archaeologists have been interested in ancient glasses as a matter of research according to the following three objectives:

1. Glass production throughout History. From raw materials localization up to the melting process of glasses, as well as some components added as stabilizers or colouring agents, the latter also named chromophores<sup>1-6</sup>.
2. Tracing of possible trade routes, either on raw materials or on finished items<sup>7-12</sup>.
3. Degradation mechanisms experienced by ancient glasses as a consequence of their contact with different environmental agents, thereby improving restoration and conservation techniques<sup>13-18</sup>.

Date of publication, journal and article types, topic, glass typology, analytical techniques, origin country of authors, and geographic location of samples were analyzed, among other parameters, in this bibliometric study. A representative set of international journals and congresses were covered. In addition, some significant Spanish examples of both categories

were also revised due to the nationality of authors who sign the present paper. This point has additionally served to assess the evolution of this kind of studies in the Spanish research community.

## 2. METHODOLOGY

To carry out the study, the main historical-archaeological and scientific journals, both Spanish and international, were consulted (Tab. I). The articles chosen were those that accomplished an archaeometric study on historical and/or archaeological glass materials. Neither works on ceramic glazes and vitreous pastes nor descriptive articles without an analytical content were taken into account. Additionally, and in order to obtain a more completed picture on the role of historical glasses into the current scientific arena, the proceedings of the following two congresses were also consulted: *International Congress on Glass* and *Congreso Ibérico de Arqueometría* (Iberian Congress on Archaeometry). Certainly, there are other congresses in which this kind of contributions are also published but, on a general basis, they were considered with less significance to draw an evolution as that studied in this paper.

The bibliometric study was focused on the last 21 years, since 1987 to 2008. The year of 1987 was chosen as starting date due to the number of articles published before this year was scarce and barely relevant. Yet, as it will be established later, the time period selected is wide and representative enough to accurately assess the developments and trends recently occurred in this research field.

## 3. RESULTS AND DISCUSSION OF THE BIBLIOMETRIC STUDY

### 3.1. Publication date

This parameter addressed the evolution experienced by archaeometric studies on glasses in the last 21 years (Fig. 1). A noticeable increment in the number of articles can be appreciated, which indicates that this research line is certainly growing and expanding. In Figure 1, two anomalies are

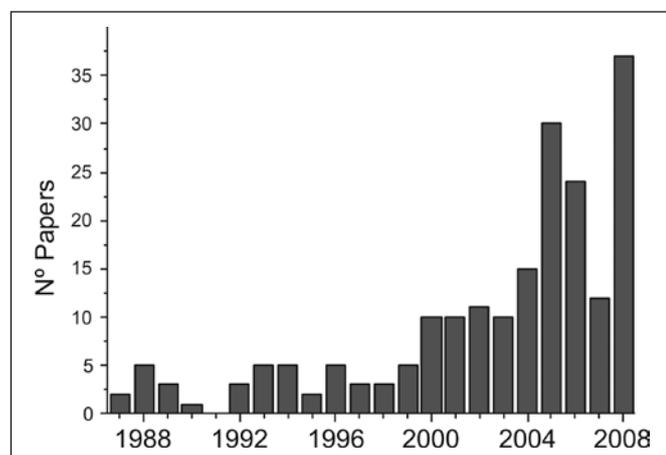


Figure 1. Time scale of the number of publications on archaeometric studies of glasses.

observed. The first one corresponds to the year 2005, with a number of publications higher than expected; and the second one to the year 2007, with a lower number. The decreasing of publications in 2007 can be produced by different reasons, such as delay of late 2007 publications, which have been eventually published in early 2008. In any case, the growing trend of the last few years is clearly stated.

### 3.2. Journal type

Given the high level of interdisciplinarity existing in archaeometric studies, the type of journals in which articles are published is very diverse. Articles can be found in either historical and archaeological or experimental sciences journals, and even in journals devoted to environmental sciences. More than 50 % of the analyzed contributions are published in experimental science journals (Fig. 2a). Among them, the most outstanding are *Nuclear Instruments and Methods in Physics Research B* (IF 0.999, JCR 2008), which covers interaction of energy beams with glass materials; *Journal of Non-Crystalline Solids* (Impact Factor, IF, 1.449, Journal Citation Reports, JCR, 2008), focused on ancient and modern glasses; and *Spectrochimica Acta Part B* (IF 2.853, JCR 2008), on the application of different spectroscopies. High number of articles in experimental sciences journals is because of, in the last years

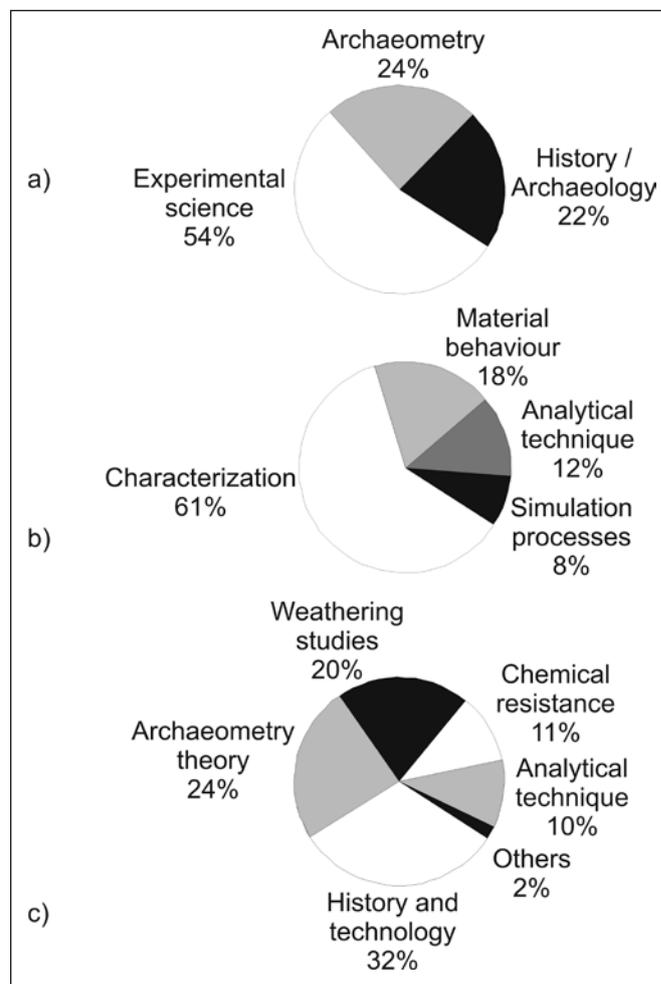


Figure 2. Percentages according to: a) journal type in which articles are published; b) different types of articles; c) topic of the article.

application of non-destructive techniques have considerably increased and, therefore, the possibilities of analysing glass samples without any damage have risen dramatically<sup>19-24</sup>. Some of these examples are published in the present journal<sup>25-28</sup>.

Despite its second general position, the journal *Archaeometry* (IF 1.479, JCR 2008) collects the highest number of articles in a single publication: 24 % (Fig. 2a). This journal is mainly focused on application of chemical-physical techniques to a wide range of historic materials, such as ceramics, bones, cloths and, certainly, glasses. Finally, in the last place history / archaeology journals are located. The most outstanding are *Journal of Cultural Heritage* (IF 0.854, JCR 2008) and *Journal of Archaeological Science* (IF 1.779, JCR 2008).

### 3.3. Article type

The study also examined the articles on the basis of the following categories: a) characterization, b) material behaviour, c) techniques and d) simulation processes (Fig. 2b).

a) *Characterization*. It covered those articles in which chemical-physical analysis are used to describe some of the glass properties, such as colour, hardness or resistance.

b) *Material behaviour*. Historical and archaeological glasses are taken to study the variation of a given property with respect of time, temperature or any other external parameter.

c) *Techniques*. Some of the most innovative analytical techniques, their physical principles, their application to model glasses and extrapolation of results to historical glass samples are covered in this section<sup>24</sup>.

d) *Simulation processes*. They are theoretical or practical studies on models to simulate degradation processes of glasses or, for instance, to determine the structural arrangement of colouring agents or chromophores within the silica network<sup>29-31</sup>.

Most of the publications consulted (61 %) lie in the *Characterization* category (Fig. 2b) and they are undertaken with the purpose to determine chemical composition of glasses, concentration of chromophores or location of possible raw materials.

### 3.4. Article topic

Articles can be also classified, according to their objectives, into the following different topics (Fig. 2c):

a) *History and Technology*. They are those articles in which technology, production and raw materials origin are studied<sup>32-35</sup>.

b) *Chemical Resistance*. They are contributions mainly focused on glass degradation processes and interaction of glasses with external agents<sup>14, 18, 36-39</sup>.

c) *Archaeometric Theory*. The articles of this topic show a glass characterization based on its chemical composition and the type of chromophore present in the glass<sup>40-44</sup>.

d) *Weathering Studies*. They are articles focused on the study of historical glasses attempting to know weathering mechanisms on their surfaces.<sup>16, 45-47</sup>

e) *Analytical Technique*. In this case, a given technique is developed to analyse model glasses, which simulate chemical compositions of ancient ones, to justify the utility of the technique. Likewise, it is also applied to historical / archaeological glasses to prove data obtained in model ones<sup>20, 21, 24, 48-53</sup>.

f) *Others*. Articles focused on other topics not included in the six previous ones, such as micro-biologic attack<sup>54, 55</sup>.

The main topic is *History and Technology*. In this case, the chemical-physical analyses provide precious knowledge on chemical composition, melting temperature and, even, on the technology existing in the time of production. The second topic with the higher amount of articles is *Archaeometric Theory*. In these works, authors resort to chemical-physical characterization techniques as a way to reconstruct ancient technology, since there exist very few written sources and documents on techniques used in the past to produce glass, such as the book *De originibus rerum* by Hrabanus Maurus (776-856) on fabrication of Medieval glasses. Finally, the third more frequent topic is *Weathering Studies*. The authors pay attention to the different mechanisms that produce degradation, the distinct reactions for which corrosion proceeds and the possible methods to avoid future damage of original glass pieces.

The rest of topics are less studied. Now because they are very specific topics, such as the *Analytical Techniques* or *Chemical Resistance*; now because topics are very innovative, such as deterioration of glasses produced by fungi, bacteria or micro-organisms, which are grouped into the *Others* topic<sup>30</sup>.

### 3.5. Type of glass

Historical and archaeological glasses may have different composition and colouring, as a result of their chronology and place of production (Fig. 3). Throughout History, glass has been used as an ornament element in the form of beads threaded in necklaces and bracelets, also as tableware items and, in the Middle Age, stained glass windows were introduced in

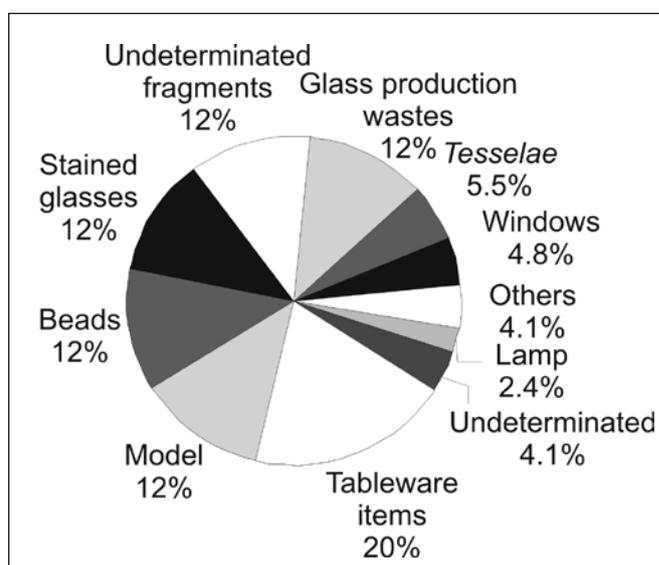


Figure 3. Typology of the glasses analyzed.

the most important European cathedrals, churches and civil buildings as a way to close big openings of the walls. It is for such reasons why this type of pieces has been more frequently studied. However, it is important to note that a considerable number of studied samples correspond to indeterminate fragments. Brittleness and relatively fast degradation of glasses determine that a lot of pieces result unrecognizable with the past of time. Thus, chemical composition of these pieces can be studied and compared with other chronologically contemporary glasses, even though it is rarely possible to assign them to a specific typology.

Another important percentage corresponds to model glasses obtained in the laboratory to carry out simulation studies<sup>56</sup>. Finally, in a low proportion, there are samples coming from glass production wastes, such as frits, crucibles, melting batches, etc.; and other objects such as lamps, *tesselae* from mosaics or flat glass from windows<sup>48, 57-59</sup>.

### 3.6. Chronology and geographic location

In Figure 4 maximum percentages coincide with Roman/Late Roman and Medieval periods. Two ages in which glass production was very important. In Roman times due to the wide diffusion of utilitarian and tableware glasses, and in the Middle Age due to the large amount of stained glass windows produced, either for religious or civil buildings. In the rest of chronological periods, the number of samples remains constant.

Geographic location of samples is directly related to the two chronological periods mentioned above. Thus, the Roman Empire, which expanded by the whole Mediterranean area, has enabled the finding of Roman glasses mainly in Europe and in the north of Africa; while in the Middle Age glasses come from the most important European cathedrals.

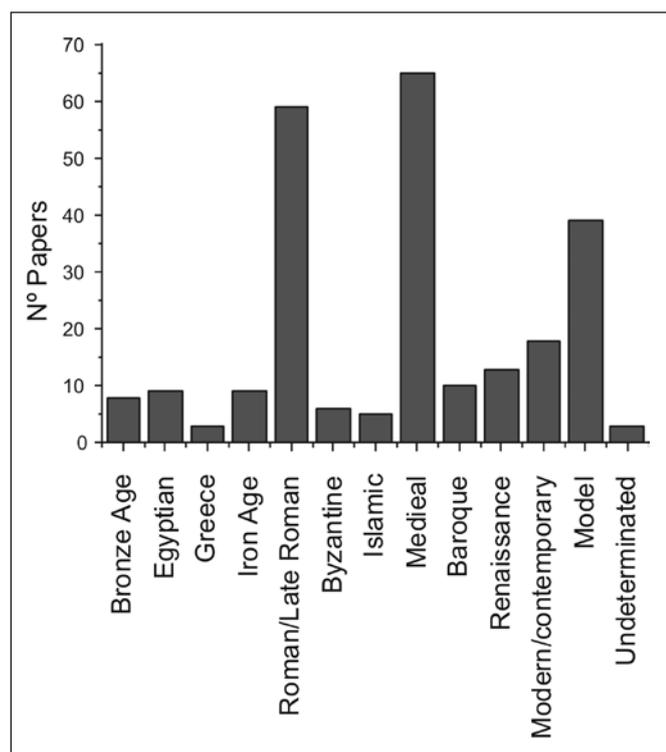


Figure 4. Chronology of glass samples analyzed in publications.

The majority of glass samples are located in Europe (59 %) (Fig. 5a), with Italy, Spain, the United Kingdom and France as principal countries (Fig. 5b). Immediately after, samples come from Asia (14 %), mainly from the Middle East. Analyzed glasses from Africa (8.9 %) are to a large extent from Egypt. As far as America is concerned, glasses come, above all, from the colonial trade. The category named *Aquatic Bottom* includes those glasses found in shipwrecks<sup>17, 47</sup>.

### 3.7. Authors

An interesting form to distribute articles consulted is the origin of the corresponding research team. As origin country, it has been taken that which appeared in the address of authors and institutions in the title page of articles. Italy is the country with the highest number of publications, mainly on Roman glasses. In the second place, it appears the United Kingdom,

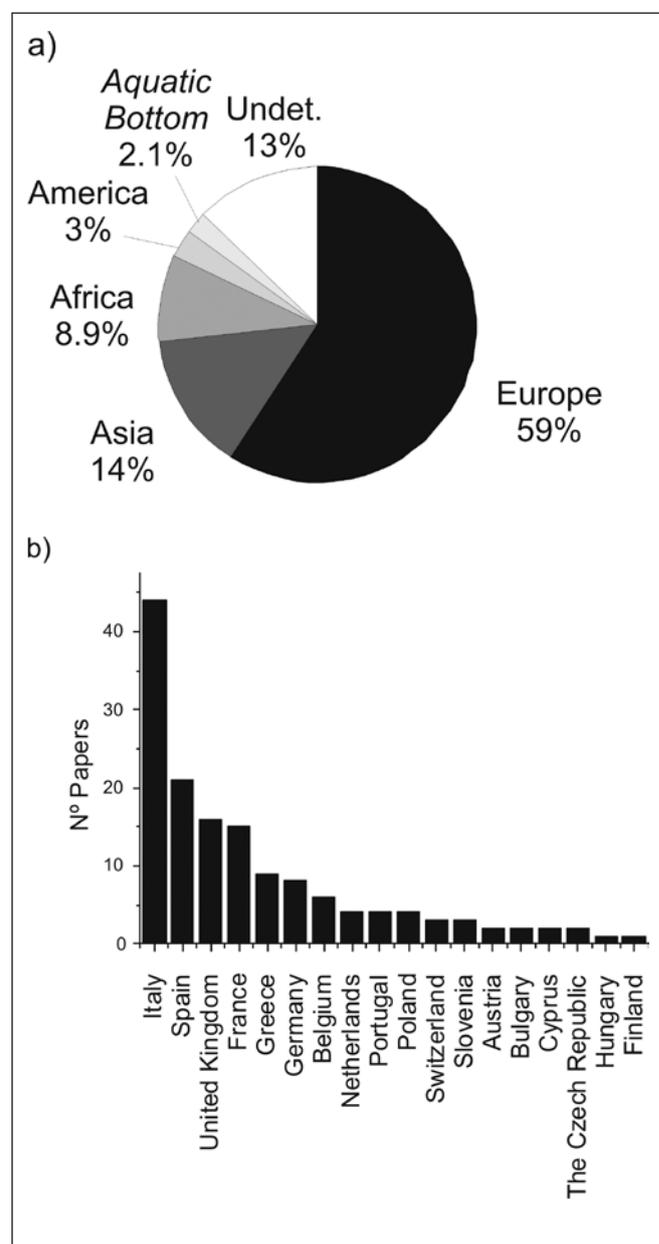


Figure 5. a) Geographical location of glass samples analyzed. b) Distribution of glass samples analyzed in European countries.

followed by Spain, Germany and France in the third, fourth and fifth places, respectively (Fig. 6).

### 3.8. Techniques used

To accomplish archaeometric analyses it is necessary an analytical instrumentation that resolves mainly both the chemical composition and the microstructure. Accordingly, the techniques used can be classified into observation techniques, to examine samples under high magnification; and analytical techniques, which are those able to determine chemical composition of glass samples. Figure 7a shows that most of the techniques used are analytical, since they are employed to know the amount of elements or oxides present in glasses. On the contrary, the observation techniques are less used.

Four different observation techniques are mainly employed: scanning electron microscopy (SEM), conventional optical microscopy (OM), transmission electron microscopy (TEM) and atomic force microscopy (AFM) (Fig. 7b). OM is widely used to observe macroscopic morphology of surface alterations, such as fissures, corrosion crusts and other surface deteriorations. SEM has been also quite used. It allows the observation of microstructure; it is non-destructive and can be attached to an energy dispersive X-ray spectrometer (EDX) to undertake chemical microanalyses in very small areas of samples. TEM and AFM techniques can be also employed to observe microstructures, even though at a higher magnification. Through AFM it is possible to distinguish individual atoms. Such techniques are not very often used perhaps due to their higher cost and reduced availability. Moreover, the information gathered is commonly from very localized areas, which limits the reproducibility and representativeness of measures.

As far as analytical techniques concern (Fig. 7c), the most often used are those employed to determine chemical composition of glasses, such as energy dispersive X-ray

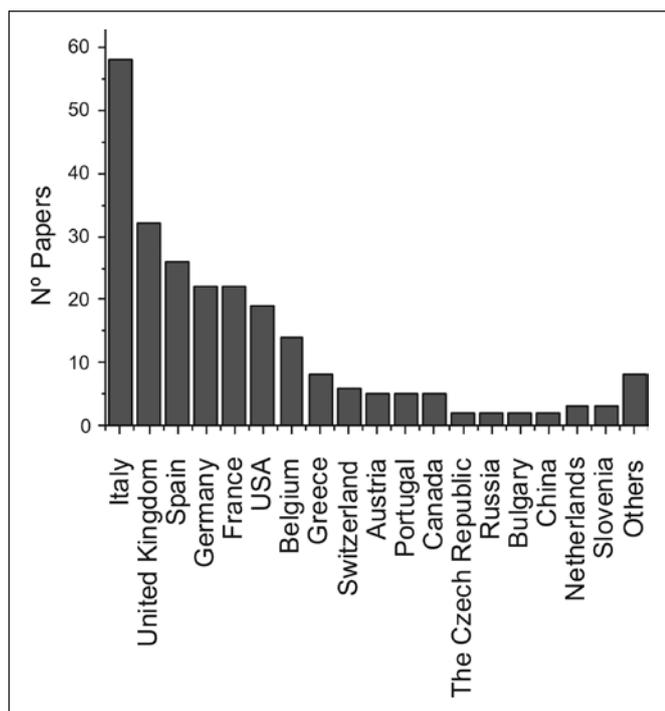


Figure 6. Distribution of origin countries of authors. Others: Australia, Bielorrussia, Finland, Hungary, Israel, Poland, Sweden and Tunisia.

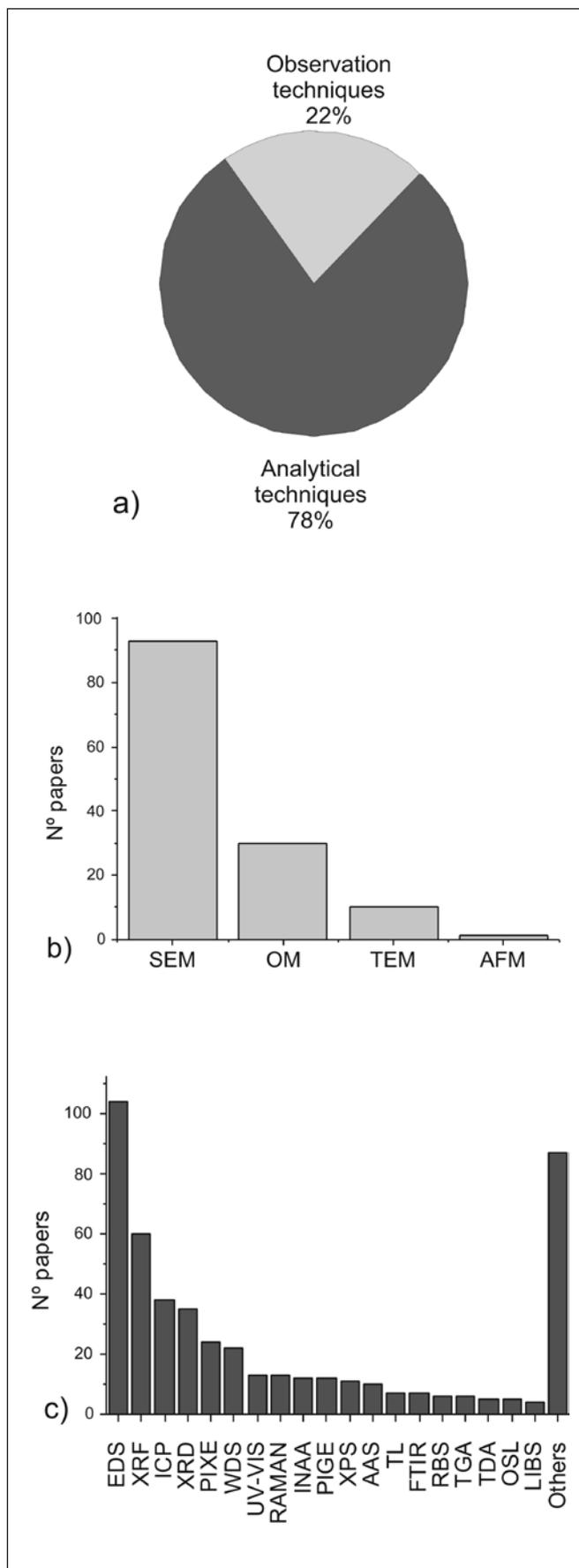


Figure 7. a) Percentage distribution of employed techniques. b) Observation techniques. c) Analysis techniques. Others: thin-sections, Mössbauer spectroscopy, Electronic Paramagnetic Resonance (EPR), etc.

(EDX), X-ray fluorescence (XRF), inductively coupled plasma (ICP), proton induced X-ray emission (PIXE), and wavelength dispersion (WDS) spectroscopies, as well as instrumental neutron activation analysis (INAA). Although in a low proportion, there are techniques that also determine other properties of glasses, such as X-ray diffraction (XRD), to identify crystalline phases in devitrified glasses; UV-Vis spectroscopy, to determine chromophores; differential thermal analysis (DTA), to assess thermodynamic changes and to determine  $T_g$  values; or thermo-luminescence (TL), to obtain information from excitation of glass samples with heat.

It is important to emphasize that the most used are the non-destructive or the partially destructive techniques, such as SEM-EDX or PIXE. However, it results still necessary to use fully destructive techniques, such as ICP or XRF, to reach lower detection limits, to determine components in very small proportions, or to improve accuracy of analyses.

### 3.9. Congresses

Two congress proceedings were reviewed: *International Congress on Glass*, which presents international scientific advances on glasses; and *Congreso Ibérico de Arqueometría* (Iberian Congress on Archaeometry), regularly held in Spain since 1995.

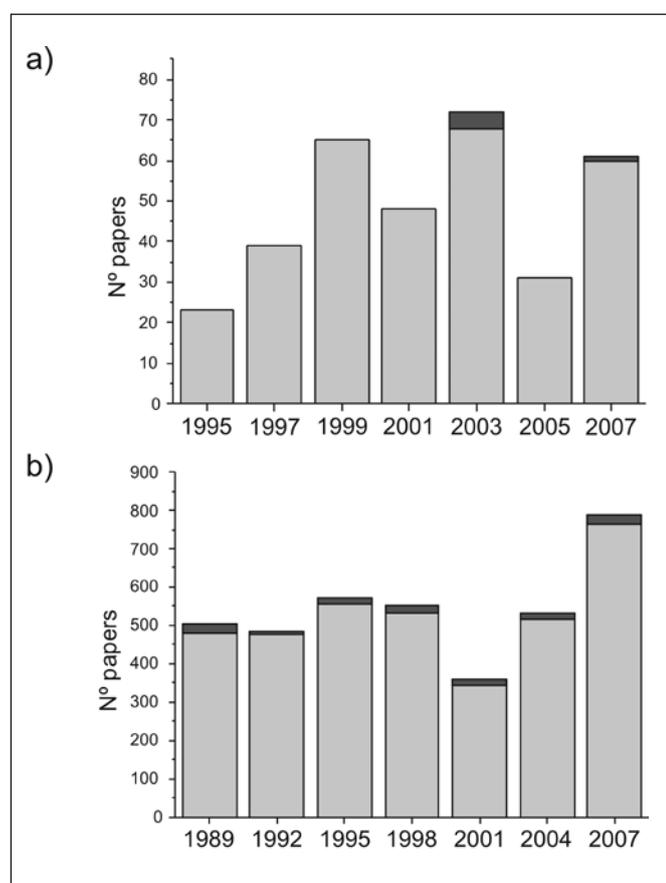


Figure 8. Distribution of contributions on archaeometry of glasses, in black, referred to the total number of works delivered to the proceedings of a) *International Congress on Glass*; b) *Congreso Ibérico de Arqueometría* (Iberian Congress on Archaeometry).

In the *International Congress on Glass* (Fig. 8a), contributions on historic and archaeological glasses have been almost constant, since there has been a session devoted to archaeometry from long time ago. Although the incidence of archaeometric contributions is lower than 5%, it is nonetheless higher than expected. It must be taken into account that this international meeting is basically devoted to scientific and technological aspects on glasses and, therefore, it is mainly focused on improvement of properties or new applications of glasses.

On the other hand, in the *Congreso Ibérico de Arqueometría* (Iberian Congress on Archaeometry), the presence of contributions referred to glasses is very scarce. Only in the 5<sup>th</sup> and 7<sup>th</sup> editions appeared a small participation, which is lower than 6% in total (Fig. 8b). The most analyzed materials are ceramics and metals; while cloths, pigments and, certainly glasses, are the materials with a lower number of contributions.

### 4. CONCLUSIONS

Results of the present bibliometric study confirm that archaeometric research on glasses with historic, archaeological and cultural interest has experienced an exponential growth during the last years (period 2000-2008). The study of the journals in which articles are published shows that only a fourth part of them, approximately, is finally edited in journals devoted to archaeometry. This fact indicates the existence of a very narrow editorial space, since the journals of archaeometry published regularly are very few.

Type of glass samples studied shows a wide variety, which corresponds to the great diversity, utility and functionality that this material got in past societies. This is a coherent aspect and can be considered as a precursor of the extensive technological development of glass in contemporary history. Most of the samples analyzed are tableware items from Roman Europe, as well as beads and ornament elements coming from Europe and the Middle East, stained glass windows from Medieval Europe and Glass production wastes. Glasses more frequently investigated belong to Roman/Late Roman and Medieval periods, since these two historical times represent significant advances in the general technological development of glass.

One of the factors influencing the increase of archaeometric studies on glass has been the development of new observation and analytical techniques, either non-destructive or partially destructive, which offer structural, microstructural and analytical information. Resulting data also indicates that analytical studies on chemical composition of glass are key approaches to establish relationships with geographical provenance of possible raw materials and weathering environmental conditions of conservation. Finally, some European countries appear as the highest producers of contributions related to glass archaeometry. Italy, the United Kingdom, Spain and Germany, respectively, occupy the first four places.

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TABLE I. LIST OF THE TOTAL NUMBER OF ARTICLES REVISIED.

Title	Paper Journal	Digital Journal	Total
Aerobiologia	0	1	1
Analytica Chimica Acta	0	1	1
Analytical and Bioanalytical Chemistry	0	1	1
Applied Clay Science	0	1	1
Applied Geochemistry	0	1	1
Applied Physics A: Materials Science & Processing	0	8	8
Applied Surface Science	0	2	2
Archaeometry	13	35	48
Boletín de la Sociedad Española de Cerámica y Vidrio	3	1	4
Chemical Geology	0	1	1
Fresenius' Journal of Analytical Chemistry	0	1	1
Geochimica et Cosmochimica Acta	0	1	1
Glass Science and Technology: Glastechnische Berichte	4	3	7
International Biodeterioration & Biodegradation	0	1	1
International Journal of Mass Spectrometry	0	1	1
International Journal of Radiation Applications and Instrumentation. Part A. Applied Radiation and Isotopes	0	1	1
Journal of Archaeological Science	0	17	17
Journal of Cultural Heritage	0	23	23
Journal of Material Science	0	3	3
Journal of Non-Crystalline Solids	0	19	19
Journal of Radioanalytical and Nuclear Chemistry	0	4	4
Journal of Raman Spectroscopy	1	1	2
Journal of the European Ceramic Society	0	1	1
Materiales de Construcción	2	0	2
Materials Characterization	0	1	1
Materials Chemistry and Physics	0	2	2
Microchemical Journal	0	1	1
Microchimica Acta	0	4	4
Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms	0	20	20
Optical Materials	0	1	1
Physica B: Condensed Matter	0	1	1
Quaternary Science Reviews	0	1	1
Radiation Measurements	0	1	1
Solid State Ionics	0	1	1
Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy	0	1	1
Spectrochimica Acta Part B: Atomic Spectroscopy	0	9	9
Surface Engineering	0	1	1
Talanta	0	1	1
Thermochimica Acta	0	2	2
Trabajos de Prehistoria	2	0	2
Zephyrus	1	0	1
	26	175	201

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