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Original article

## Mineralogical and microstructural characteristics of historic Roman cement renders from Budapest, Hungary

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#### ABSTRACT

Roman cements, one of the most extensively produced types of hydraulic binders of the second half of the 19th century, played an important role in the architecture of many European countries. This paper deals with the chemical-mineralogical and microstructural characterisation of historic Roman cement renders from Budapest, Hungary. Different microscopic techniques were used on polished thin sections and fracture surfaces in order to understand the method of producing these renders and the effect of urban pollution on them. The renders exhibited characteristics typical to a Roman cement mortar, such as high binder to aggregate ratios (b/a), mostly fine-grained aggregates and high capillary porosity, but without the shrinkage cracks that are also normally present. This research suggests that coarse residual cement grains may have acted in a manner similar to aggregates by absorbing stress and thereby reducing the formation of shrinkage cracks. Based on the mineral characteristics of residual cement grains, the samples could be divided into two groups, which correspond to either a higher or lower temperature of calcination of the original source material of the cement. Chemical characteristics of the binders suggest the presence of intermixed CaCO<sub>3</sub> originating from the carbonation of hydration products and partly from residual calcium carbonate of the raw material. Despite dense and often impermeable coats applied in later renovations and exposure to a polluted urban environment, which resulted in formation of gypsum on the surface of the renders, the samples show good to excellent state of preservation after more than a century. The strong "house of cards"-like arrangement of the complex C-(A)-S-H-type phases is responsible for both the high capillary porosity and the good resistance of Roman cement renders to atmospheric pollution and potentially damaging salts such as Na- and K-chlorides which are found near the base of the building due to sidewalk de-icing. These results help to better understand the behaviour of historic Roman cement renders, which in turn assists in making good decisions in choosing a repair material to future restorations of 19th century façades built with this material.

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#### 1. Introduction

Roman cement (RC), patented in 1796 by James Parker in England [1] and named because it shared with the lime-pozzolan binder used by the ancient Romans the ability to set hydraulically, took almost half a century to disseminate to the European continent [2]. Most of the Central European countries, such as the Austro-Hungarian Empire, imported small amounts of RC from England before 1850, around when they started their own production, using

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local raw materials [3]. One of the earliest Central European applications of RC, based on locally produced binders, took place in 1840 in Pest-Buda (now Budapest). A marlstone from Beocsin/Beočin (in what is today Serbia) was locally burnt by English engineer William Tierney Clark for use in the construction of the piers of the Chain-Bridge crossing the Danube River [4,5]. In the second half of the 19th century, the Austro-Hungarian Empire became the largest producer of RC in Central Europe, where until the end of the 19th century, the major part of the cement production was RC; Portland cement (PC) was a hydraulic binder of secondary importance [5]. At the turn of the 20th century, the production of PC increased rapidly while the importance of RC declined, due in part to a change in style of architecture towards modern functionalism with an absence of ornaments. The more consistent properties of PC due to the standardisation of production technology [2] also played an important role.

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coatings of later renovations

Fig. 1. Characteristic apartment house in the Budapest downtown with slightly damaged diamond-pointed rustication at the plinth made of Roman cement (RC-2).

Roman cement is a highly hydraulic natural cement formed by calcining marls, a fine-grained sedimentary rock that naturally contains carbonate and silicate components in intimate mixture, below the sintering temperature, i.e. between 800 and  $1100 \circ C$  [2,6,7]. Marls were traditionally burnt in vertical shaft kilns where, due to the low burning temperature, coarse raw feed and large temperature gradients, clinkers with varying degrees of calcination were formed. Based on clinker analyses carried out on a number of historic RC mortars and of systematic calcination tests in laboratory, it is possible to create a RC that is a close match to those created in the 19th century [2,6].

Restoration of historic façades is one of the major tasks in protection of architectural cultural heritage. As a part of the ROCARE project, supported by the 7th Framework Programme of the European Commission, a statistical research aiming at detecting the percentage of 19th century historic buildings in Budapest with façades built of RC renders has been carried out [8]. Being aware of exact historic data regarding the nature and use of RCs in Hungary, the architectural-statistical research has been completed with the material analysis of several historic mortar and render samples. This paper reports a study concerning the chemical-mineralogical composition, microstructure and overall condition of a selection of RC renders. The information gained from this investigation helps to



**Fig. 3.** Sample RC-3: fine-grained aggregates (white) and coarse, differently fired cement grains (slightly overfired with  $C_2S$  clusters: center right; optimally fired: dark, subrounded to rounded relics partly with hydration rims: center bottom and top; parallel polars).

better understand the behaviour of historic RC renders and can be used to create a compatible material for use in restoring rendered façades [9,10].

#### 2. Sampling and analytical methods

Seven multi-layered render samples containing the original RC renders along with the layers of later renovation or construction phases (Fig. 1) were taken from the façades of apartment buildings constructed between 1870 and 1912 (Table 1). Polished thin sections were prepared from the samples that had been impregnated under vacuum with a low viscosity epoxy resin dyed blue. They were analysed with optical microscopes (Nikon SMZ 1500, Zeiss AXIOScope A1) using both transmitted bright and dark field as well as incident light. The binder-to-aggregate ratio (b/a) and the grain-size distribution of aggregates and cement remnants were estimated for each sample by using a polarising microscope at magnifications  $50-100 \times$ , by superimposing a square grid and area counting [11]. A scanning electron microscope (SEM, Zeiss EVO MA 15) coupled with an energy dispersive X-ray spectrometer (EDS, Oxford DryCool) was used to make a number of point analyses



**Fig. 2.** Roman cement mortar covered with several lime-, PC- and acrylic dispersionbased coatings (RC-1; bright field of view, parallel polars: BF and dark field of view: DF).



**Fig. 4.** Coarse residual cement grains (dark and bright brown particles) and finegrained, well-sorted aggregates (white) in the sample RC-6 (bright field of view, crossed polars: BF; incident light: IL).

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