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Investigations of Physical and Chemical Characteristics of Masonry Stones and Bricks during Building Cleaning: Part 2. Chemical Testing

Binsheng Zhang[1], Humayun Reza[2], Shiyan Gu[2] and Naren Gupta[2]

1. School of Engineering and Built Environment, Glasgow Caledonian University, Cowcaddens Road, Glasgow G40BA, United Kingdom

2. School of Engineering and the Built Environment, Merchiston Campus, Edinburgh Napier University, 10 Colinton Road, Edinburgh EH10 5DT, United Kingdom

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Abstract: This series of study focused on analysing and assessing the changes of the physical and chemical characteristics of the stone surfaces during the sandblasting cleaning process by conducting various physical and chemical tests. Seven masonry stones and bricks were adopted, including yellow sandstone, red sandstone, limestone, marble, granite, white clay brick and yellow clay brick. The chemical investigations included the micrographing of the stone façade and the analysis of the chemical elements and compounds on four of the seven stones and bricks before and after the cleaning using the Scanning Electron Microscope (SEM) and Energy-Dispersive X-ray Spectroscopy (EDX) techniques. In general, the chemical properties were found to vary largely during the building cleaning. The chemical tests showed that the chemical elements and compounds on the stone façade significantly varied after long term exposures to the atmosphere, mainly due to the soiling on the building façade caused by environmental erosion and weathering.

Key words: Masonry stone and brick, sand blasting cleaning, chemical analysis, SEM, EDX.

1. Introduction

Masonry stones and bricks have been widely used for constructing historic buildings and monuments, which become grand assets for current and future generations. The cleaning and restoration of these old, historic stone and brick structures has also become significantly important accordingly. With the development of new building legislations and modern cleaning techniques in the past few decades, building cleaning nowadays has become a less aggressive practice and a more popular business [1-6]. In the United Kingdom, large demands of stone cleaning have occurred since [7-9]. Also, more attention has been paid to this and many studies on building cleaning have been published [10-18].

Frankly speaking, stone cleaning no matter how big care is taken always has negative effects beyond the removal of superficial soiling. When carried out using inappropriate methods, aggressive cleaning can largely damage stones. Many of the potential effects of inappropriate cleaning will be visible immediately after or within a few weeks of cleaning.

Hence, preliminary investigations on both physical and chemical characteristics of the masonry stone and brick surfaces are sometimes needed before deciding on the best cleaning method to avoid unnecessary damage to the buildings [10, 19-21]. However, so far there are no consistent standards and parameters used for assessing the degree of building cleaning, and the

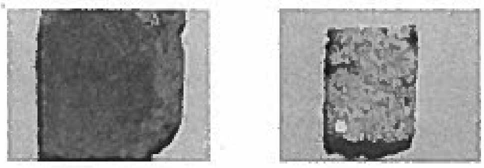
Corresponding author: Binsheng Zhang, Ph.D., MSc, B. Eng, senior lecturer, research field: civil and structural engineering. E-mail: Ben.Zhang@gcu.ac.uk.

efficiency of various cleaning methods is largely assessed by visual inspections and mutual agreements. There is an urgent need to search for better physical parameters for such assessments. Previous studies were largely focused on finding the substances of the soiling on the building façade and the methods to remove these substances. The information on the chemical compositions of the soiling and their changes during masonry cleaning is still limited. Meanwhile there is a lack of systematic monitoring and assessment on the changes in the physical and chemical characteristics of masonry stones and bricks during cleaning process even though such knowledge is largely important for understanding and improving the efficiency of building cleaning.

In this study, physical and chemical characteristics of masonry stones and bricks subjected to progressive stages of cleaning were investigated for evaluating the effectiveness of building cleaning. Part 1 of this study had reported the physical tests including digital image analysis method based on surface greyscale, hardness tests and water absorption tests [22]. Seven types of commonly used masonry stones and bricks selected for physical tests were yellow sandstone, red sandstone, limestone, marble, granite, white clay brick and yellow clay brick. This second part of the work would report the chemical analysis carried out to quantitatively assess the variations of chemical elements on the original dirty and fully clean surfaces of the masonry stones and bricks using combined Scanning Electron Microscope (SEM) and Energy-Dispersive X-ray Spectroscopy (EDX) techniques to identify the chemical compositions of the soiling on the masonry surface. Four out of the seven masonry stones and bricks selected for the physical tests were adopted for the chemical analysis, including yellow clay brick, yellow sandstone, limestone and marble. Thus, a complete evaluation procedure for building cleaning can be established.

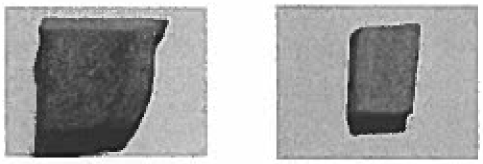
2. Preparation of Stone Samples

Masonry stones and bricks were selected from those for the 1860s-1870s listed buildings in the south west of the city of Edinburgh, which were popularly used for local buildings [23] and exposed to the open environmental conditions for more than a century with large amounts of heavy soiling deposited on the surfaces. A diamond saw was used to cut the masonry stones into small samples. The exposed surfaces of the stones and bricks were cleaned into different levels using the abrasive sandblasting cleaning with fine recycled glass particles, and then they were cut into the required sizes for various physical and chemical tests. Figs. 1 to 4 show the fully dirty and fully clean samples of yellow clay brick, yellow sandstone, limestone and marble for chemical analysis.



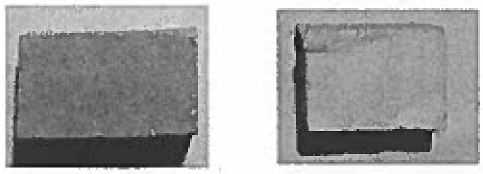
(a) (b)

Fig. 1 Yellow clay brick samples for SEM and EDX testing. (a) fully dirty sample and (b) fully clean sample.



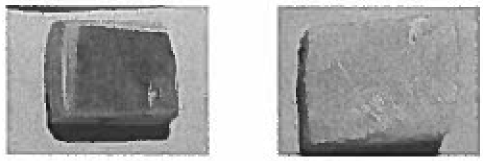
(a) (b)

Fig. 2 Yellow sandstone samples for SEM and EDX testing: (a) fully dirty sample and (b) fully dirty sample.



(a) (b)

Fig. 3 Limestone samples for SEM and EDX testing: (a) fully dirty sample and (b) fully dirty sample.



(a) (b)

Fig. 4 Marble samples for SEM and EDX testing: (a) fully dirty sample and (b) fully dirty sample.

3. Chemical Analysis

As the soiling and decay have the ability to affect the chemical substances on the stone or brick surface, the chemical characteristics of the original dirty surface are largely different to those on the fully clean surface. During the cleaning process, the chemical substances on the stone or brick surface continually change. Some chemical elements and compounds may increase and some elements and compounds may decrease or even disappear during building cleaning. This part of the work aimed to conduct quantitative chemical analysis on changes in chemical elements and compounds on the original dirty and fully cleaned (fresh) surfaces of masonry stones and bricks during cleaning process and to identify the chemical compositions and compounds of the soiling deposited on the stone and brick surfaces so as to find appropriate cleaning methods.

So far chemical analysis has been largely used for detecting the chemical compositions and compounds of the soiling remaining on masonry historic buildings and monuments after years' weathering, environment erosion and industrial pollutions [24, 25]. It is also largely used for assessing the performance of stone protection methods for conservations of historic buildings and monuments [26-29].

Most popularly used chemical analysis methods include SEM and EDX. The SEM technique is used to image a sample on a Liquid Crystal Display (LCD) by scanning it with a beam of electrons in a raster scan pattern. This will produce the signals containing the information about the surface topography and composition of the sample due to the interactions between the electrons and atoms. The EDX is used to analyse the chemical elements and compounds of the sample, based on an interaction of the source of X-ray excitation with a masonry sample. Its characterisation capabilities are largely due to the fundamental principle that each element has a unique atomic structure allowing a unique peak on its X-ray spectrum. It will be possible to detect the chemical elements on the different parts of the sample, and these elements can be related to certain chemical compounds.

In this study, the chemical analysis was conducted by using the instrument with the combined SEM and EDX, as shown in Fig. 5. The instrument used in this study was the SEM LEO S 430 I, UK, coupled with ISIS EDD detector from Oxford Instrument, UK.

Sample preparation is a vital stage for the testing using the Scanning Electron Microscope. Insulation materials are required to form a thin layer of conducting coating (~100 Å) to avoid charging. For the EDX in this study, carbon coating was adopted. The materials could be observed at low primary energy, at which the coefficient for secondary emission was ~1 and the charge build-up was negligible. The entire sample preparation included mounting the sample on a metallic platform via a conducting path.

Four adopted masonry stones and brick to be tested were numbered as:

- Yellow clay brick: Samples 1 (original dirty) and 2 (fully clean);
- Yellow sandstone: Samples 3 (original dirty) and 4 (fully clean);
- Limestone: Samples 5 (original dirty) and 6 (fully clean);
- Marble: Samples 7 (original dirty) and 8 (fully clean).

The surfaces of the fully clean samples were polished and cleaned using acetone. The original dirty samples were also gently rinsed using acetone. All the

Fig. 5 The SEM and EDX instrument.

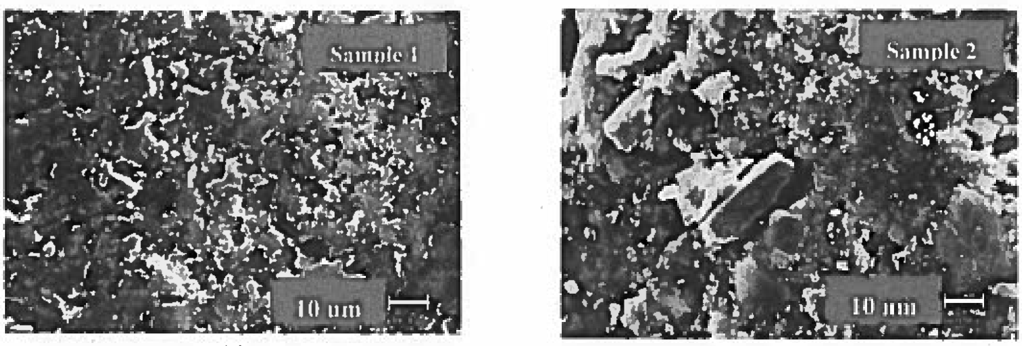
samples were dried under an IR lamp and coated with a thin layer of carbon to make the stone surfaces conductive. The samples were then mounted on the SEM stubs for the micro-structural and compositional analysis. Six micrographs were recorded at different magnifications for each sample by using the SEM and six sampling points were selected on each sample for detecting the chemical elements and compounds.

4. Yellow Clay Brick

Fig. 6 presents typical micrographs of the surface structures of the original dirty and fully clean yellow clay brick samples. Fig. 6a shows that the soling existed loosely on the dirty surface, and there were no obvious interactions between the particles. Fig. 6b shows that the fully clean surface was more crystalline and interactive. The numbers in the brackets represent the sampling points on the sample.

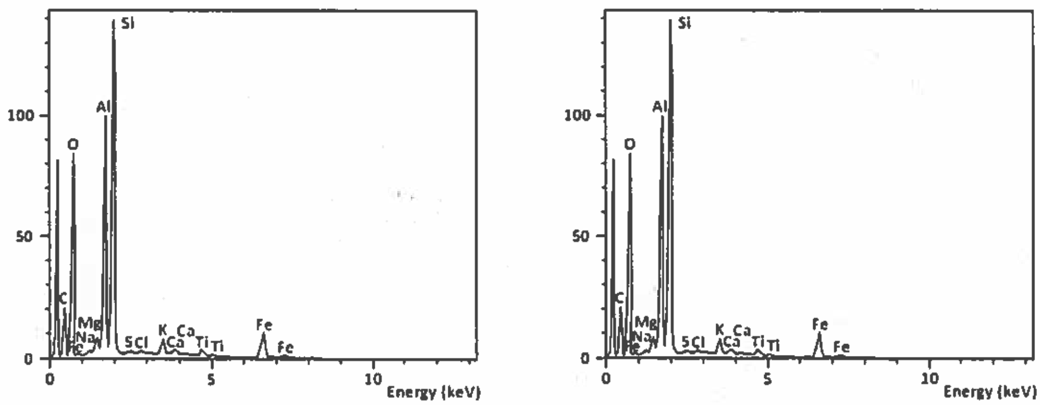
Fig. 7 shows typical chemical spectrum diagrams on the original dirty and fully clean surfaces of the yellow clay brick samples. Common chemical elements found to exist on both dirty and clean surfaces included C, O, Na, Mg, Al, Si, S, Cl, K, Ca, Ti and Fe, but the peak values were remarkably different for some elements, e.g. C, Al, Si, S, Ca and Fe, which indicates that the amounts of these elements varied during the cleaning process.

Table 1 lists the relative amounts of these thirteen detected chemical elements in percentage obtained by using the EDX for both original dirty and fully clean yellow clay brick samples. These values were the averages of six test results for each sample. The standard deviations (SD) for each chemical element are also included in the table. Compared with the average



(a) (b)

Fig. 6 Typical micrographs for the yellow clay brick samples. (a) Original dirty surface (Sample 1(6)) and (b) fully clean surface (Sample 2(5)).



(a) (b)

Fig. 7 Typical spectrum diagrams for the yellow clay brick samples. (a) Original dirty surface (Sample 1(5)) and (b) fully clean surface (Sample 2(4))

Table 1 EDX results for the yellow clay brick samples.

SEMQuant results			Ref: Demonstration data SiLi detector				Spectrum label: Samples 1&2
System resolution = 61 eV			Quantitative method: ZAF (6 iterations)				Analysed all elements
Element	Spectrum type		Element (%)				Chemical compound
			Dirty		Clean		
			Average	SD	Average	SD	
C	K	ED	23.50	2.19	28.80	9.58	$CaCO_3$ 01/12/93
O	K	ED	45.26	0.80	45.80	2.45	Quartz 01/12/93
Na	K	ED	0.39	0.25	0.14	0.02	Albite 02/12/93
Mg	K	ED	0.55	0.16	0.44	0.10	MgO 01/12/93
Al	K	ED	8.97	1.27	4.39	1.75	Al_2O_3 23/11/93
Si	K	ED	16.42	2.65	14.12	7.54	Quartz 01/12/93
P	K	ED			0.30	0.22	GaP 29/11/93
S	K	ED	0.09	0.03	0.28	0.15	FeS_2 01/12/93
Cl	K	ED	0.22	0.09	0.18	0.14	KCl 15/02/94
K	K	ED	1.28	0.21	0.93	0.30	MAD-10 02/12/93
Ca	K	ED	0.41	0.25	2.42	1.82	Wollas 23/11/93
Ti	K	ED	0.44	0.20	0.47	0.23	Ti 01/12/93
Fe	K	ED	2.49	1.30	2.04	0.22	Fe 01/12/93
Total			100.00		100.00		

values, the standard deviations were reasonably small so the average values can be regarded to represent the true relative quantities of chemical elements on the surfaces of the yellow clay brick in this study. Also based on these quantities together with the measured atomic weights, the possible chemical compounds could be indicated, see the last column of Table 1.

Fig. 8 shows the quantities of the chemical elements detected on the original dirty and fully clean surfaces of the yellow clay brick samples. The main chemical elements in the original yellow clay brick were C, O, Si and Al at 23.50%, 45.26%, 16.42% and 8.97%, respectively, which indicates that the main chemical compounds in the yellow clay brick were $CaCO_3$, SiO_2 and Al_2O_3. By viewing the 50% dividing line, it can also be seen that that C slightly increased to 28.80% after cleaning while Si and Al decreased to 14.12% and 4.39%. As the samples were coated with carbon, it is hard to quantitatively analyse the changes of C. However, the decrease in Si and Al which represent Quartz (SiO_2) and Aluminium oxide (Al_2O_3) through the cleaning process indicates that these two compounds were formed in the original yellow clay brick. Similarly, the decrease of the rare elements in the yellow clay brick such as Mg and Fe which represent Magnesium oxide (MgO) and Iron disulfide (FeS_2) may be caused by polluting gases like O_3 and H_2S.

Punmia et al. [30] claimed that the main chemical compositions in clay bricks included 50%-60% silica (SiO_2), 20%-30% alumina (Al_2O_3), 5-6% iron oxide (Fe_2O_3), 2%-5% lime (CaO) and magnesia (MgO) below 1%. The current results seemed indeed to match

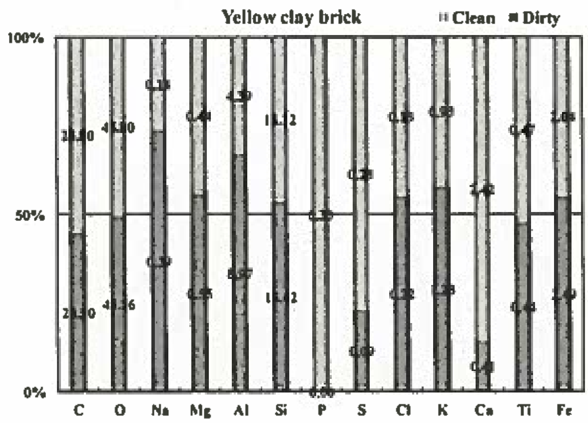


Fig. 8 Chemical elements on the surfaces of the original dirty and fully clean yellow clay brick samples.

the reported distributions. For the yellow clay brick samples in this study, the detected extra chemical elements included Na, P, S, Cl, K, Ti and their compounds which existed in both the soiling and on the fully clean surface except P.

5. Yellow Sandstone

Fig. 9 presents typical micrographs of the surface structures of the original dirty and fully clean yellow sandstone samples. Fig. 9a shows that the soling still loosely existed on the surface of the dirty yellow sandstone, and there were no obvious interactions between the particles. Fig. 9b shows that the surface of the fully clean yellow sandstone was remarkably crystalline and orderly.

Fig. 10 illustrates typical chemical spectrum diagrams on the surfaces of the original dirty and fullyclean yellow sandstone samples. Common chemical elements observed on both dirty and clean surfaces included C, O, Mg, Al, Si, K, Ca and Fe, and the peak values were remarkably different for some elements, e.g. C, Al, K, S, Ca and Fe, which indicates that the amounts of these elements varied during the cleaning process. Si and Cl only existed on the original dirty surface while Na and Ti only existed on the fully clean surface.

Table 2 lists the relative amounts of these twelve detected chemical elements in percentage obtained by using the EDX for both original dirty and fully clean yellow sandstone samples. The standard deviations(SD) for each chemical element are also included in the table. Similarly, the standard deviations were reasonably small compared with the average values, so the average values can represent the true relative quantities of chemical elements on the surfaces of the yellow sandstone.

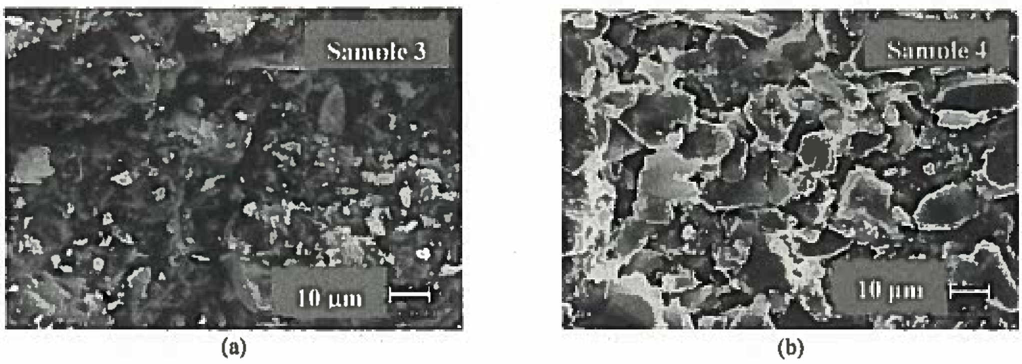


Fig. 9 Typical micrographs for the yellow sandstone samples: (a) original dirty surface (Sample 3(4)) and (b) fully clean surface (Sample 4(5)).

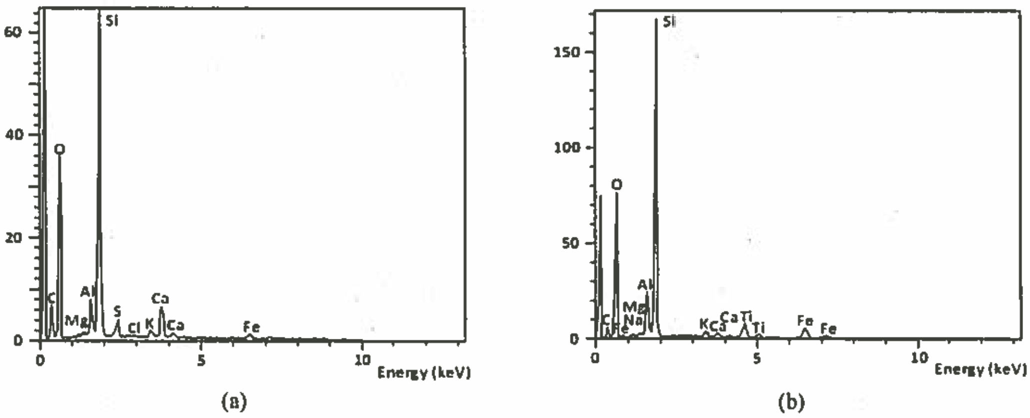


Fig. 10 Typical spectrum diagrams for the yellow sandstone sample: (a) original dirty surface (Sample 3(6)) and (b) fully clean surface (Sample 4(6)).

Table 2 EDX results for the yellow sandstone samples.

SEMQuant results			Ref: Demonstration data SiLi detector				Spectrum label: Samples 3&4
System resolution = 61 eV			Quantitative method: ZAF (6 iterations)				Analysed all elements
Element	Spectrum Type		Element (%)				Compound
			Dirty		Clean		
			Average	SD	Average	SD	
C	K	ED	19.43	3.39	13.10	1.22	$CaCO_3$ 01/12/93
O	K	ED	54.45	3.88	53.51	3.84	Quartz 01/12/93
Na	K	ED			0.25	0.05	Albite 02/12/93
Mg	K	ED	0.20	0.03	0.13	0.11	MgO 01/12/93
Al	K	ED	2.24	1.96	3.67	2.12	Al_2O_3 23/11/93
Si	K	ED	21.58	5.10	24.67	4.46	Quartz 01/12/93
S	K	ED	0.49	0.62			FeS_2 01/12/93
Cl	K	ED	0.04	0.01			KCl 15/02/94
K	K	ED	0.68	0.62	0.43	0.23	MAD-10 02/12/93
Ca	K	ED	1.14	1.12	0.85	0.72	Wollas 23/11/93
Ti	K	ED			1.36	1.75	Ti 01/12/93
Fe	K	ED	0.92	0.38	2.18	0.96	Fe 01/12/93
Total			100.00		100.00		

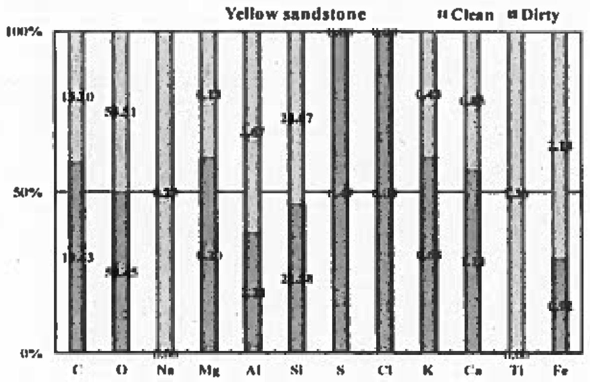


Fig. 11 Chemical elements on the surfaces of the original dirty and fully clean yellow sandstone samples.

Fig. 11 shows the quantities of the chemical elements detected on the original dirty and fully clean surfaces of the yellow sandstone samples. The mainelements in the clean yellow sandstone were C, O and Si at 13.10%, 53.51% and 24.67%, respectively, and the corresponding compounds were $CaCO_3$ and SiO_2. By viewing the 50% dividing line, it can also be seen that the main elements in the sandstone did not change much during cleaning.

However, some metallic elements such as Na, Al, Ti and Fe which represent Albite, Aluminium oxide (Al_2O_3), Titanium (Ti) and Iron disulfide (FeS_2) largely increased after cleaning, which indicates that these elements were the original elements of the yellow sandstone. The biological soiling on the stone surface such as bacteria which has the ability to largely dissolve a range of components of the stone may lead to the loss of these compounds on the original stone. On the contrast, the decrease of Mg, S and Cl which represent Magnesium oxide (MgO), Iron disulfide (FeS_2) and Potassium chloride (KCl) through the cleaning indicates that these compounds were the naturally formed soiling on the façade of sandstone, probably due to the reactions with the polluting gases such as O_3, SO_2 and H_2S in the atmosphere.

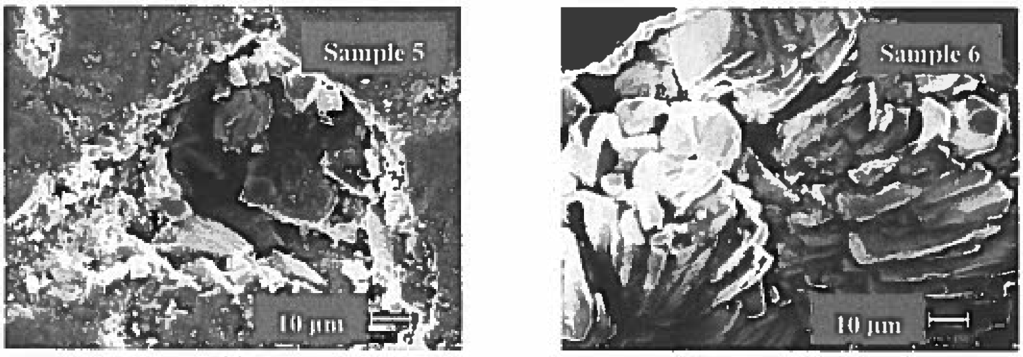
Mineral Zone [31] reported that the main chemical compositions in sandstone included 95%-97% silica (SiO_2), 1.0%-1.5% alumina (Al_2O_3), 0.5%-1.5% iron oxide (Fe_2O_3), soda (Na_2O) and potash (Kro) below 1%, lime (CaO), magnesia (MgO) and loss on ignition (LOI) below 0.5% each. The current results seemed to match the reported distributions. For the yellow sandstone samples in this study, the detected extra chemical elements included Na, S, Cl, K, Ti and their compounds, but only S and Cl existed in the soiling and Na and Ti only on the fully clean surface.

6. Limestone

Fig. 12 shows typical micrographs of the surface structures of the limestone samples. Fig. 12a shows that the soling on the surface of the dirty limestone was lightly crystalline with some defects. Fig. 12b shows that the surface of the fully clean limestone was more crystalline and orderly.

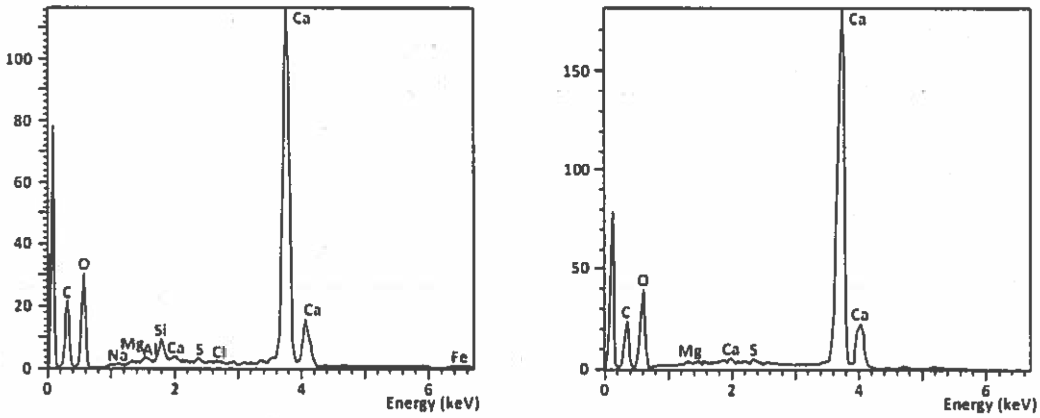
Fig. 13 illustrates typical chemical spectrum diagrams on the surfaces of the original dirty and fully clean limestone samples. Common chemical elements observed on both dirty and clean surfaces included C, O, Mg, Si and Ca, but the peak values were remarkably different for C and Ca, which indicates that the amounts of these two elements largely varied during the cleaning process. Na, Al and Si only existed on the original dirty surface.

Table 3 lists the relative amounts of the eight detected chemical elements by using the EDX for both original dirty and fully clean limestone samples. Fig. 14 shows the quantities of the chemical elements detected on the original dirty and fully clean surfaces of the limestone samples. The main chemical elements in the clean limestone were C, O and Ca at 12.80%, 49.92% and 36.87%, and the corresponding compounds were $CaCO_3$, SiO_2 and Wollas. By viewing the 50% dividing line, it can also be seen that the main elements in the limestone did not change largely during the cleaning. However, some rare elements such as Na, Al and Si which represent Albite, Aluminium oxide (Al_2O_3) and Quartz (SiO_2) disappeared after cleaning, which indicates that these compounds were not the original elements of the limestone but belonged to the dirty soiling.



(a) (b)

Fig. 12 Typical micrographs for the limestone samples. (a) Original dirty surface (Sample 5(2)) and Fully clean surface (Sample 6(4)).



(a) (b)

Fig. 13 Typical spectrum diagrams for the limestone samples. (a) Original dirty surface (Sample 5(6)) and (b) Fully clean surface (Sample 6(4)).

Table 3 EDX results for the limestone samples.

SEMQuant results			Ref: Demonstration data SiLi detector				Spectrum label: Samples 1&2
System resolution = 61 eV			Quantitative method: ZAF (6 iterations)				Analysed all elements
Element	Spectrum Type		Element (%)				Compound
			Dirty		Clean		
			Average	SD	Average	SD	
C	K	ED	15.91	1.36	12.80	0.79	$CaCO_3$ 01/12/93
O	K	ED	50.68	1.79	49.92	1.86	Quartz 01/12/93
Na	K	ED	0.29	0.13			Albite 02/12/93
Mg	K	ED	0.24	0.05	0.26	0.11	MgO 01/12/93
Al	K	ED	0.21	0.09			Al_2O_3 23/11/93
Si	K	ED	0.53	0.47			Quartz 01/12/93
S	K	ED	0.21	0.05	0.21	0.05	FeS_2 01/12/93
Ca	K	ED	32.21	3.35	36.87	1.42	Wollas 23/11/93
Total			100.00	100.00	100.00	100.00	

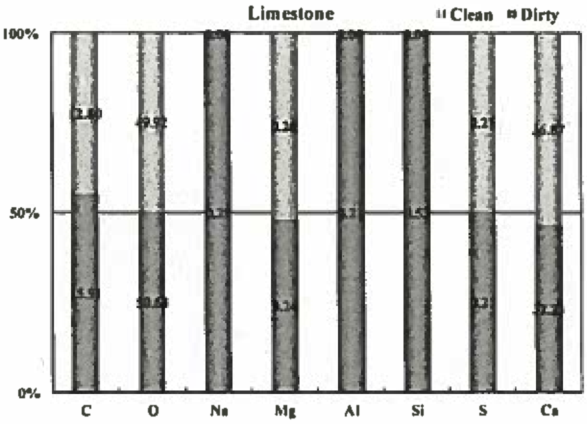


Fig. 14 Chemical elements on the surfaces of the original dirty and fully clean limestone samples.

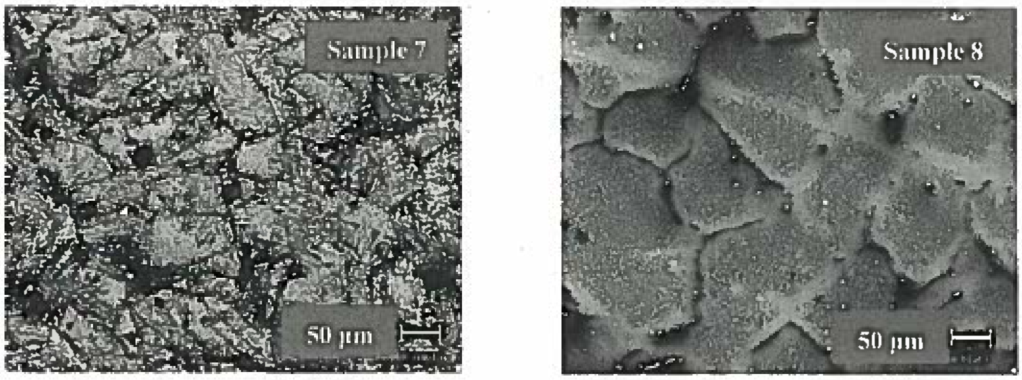
Mineral Zone [31] reported that the main chemical compositions in limestone included 15%-18% silica (SiO_2), 1%-1.5% iron oxide ($FeO + Fe_2O_3$), 38%-42% lime (CaO), 0.5%-3% magnesia (MgO), 1%-1.5% alumina (Al_2O_3), 1%-1.5% alkalies and 30-32% loss on ignition (LOI). For the limestone samples in this study, the detected amounts of lime (CaO) and magnesia (MgO) seemed to be reasonably within the reported range. Silica (SiO_2) and alumina (Al_2O_3) only appeared in the soiling on the original dirty surface but disappeared on the fully clean surface. Iron oxide ($FeO + Fe_2O_3$) did not appear on the fully clean surface at all. The extra chemical elements detected were Na, S and their compounds, and Na only appeared in the soiling on the original dirty surface but not on the fully cleaned surface.

7. Marble

Fig. 15 presents typical micrographs of the surface structures of the original dirty and fully clean marble samples. Fig. 15a shows that the soling on the surface of the dirty marble was rough and loose, while Fig. 15b shows that the surface of the fully clean marble was crystalline and orderly. Fig. 16 shows typical chemical spectrum diagrams on the surfaces of the original dirty and fully clean marble samples. Common chemical elements observed on both dirty and clean surfaces included C, O, Mg, Al, Si and Ca, but the peak values were remarkably different for C, O, Al, Si and Ca, which indicates that the amounts of these elements largely varied during the cleaning process. Na, S, Al, K and Fe only existed on the original dirty surface.

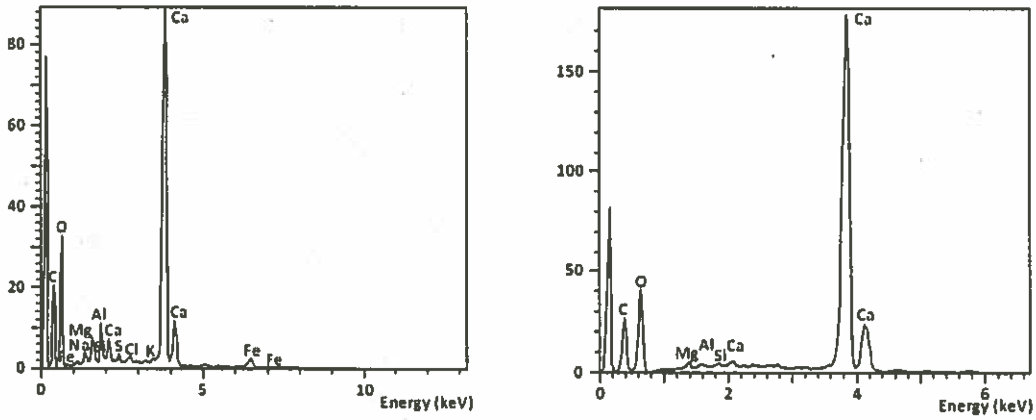
Table 4 lists the relative amounts of the eleven detected chemical elements in percentage by using the EDX for both original dirty and fully clean marble samples. Fig. 17 shows the quantities of the chemical elements detected on the original dirty and fully clean surfaces of the marble samples. The main elements in the clean marble were C, O and Ca at 12.70%, 51.27% and 35.49%, respectively, and the main compounds in the marble were $CaCO_3$ and Wollas.

It can also be seen that the rare compounds in the marble were all largely decreased after cleaning, which indicates that the surface condition of the original marble was poor as large amounts of soiling formed on



(a) (b)

Fig. 15 Typical micrographs for the marble samples. (a) Original dirty surface (Sample 7(3)) and (b) Fully clean surface (Sample 8(5))



(a) (b)

Fig. 16 Typical spectrum diagrams for the marble samples: (a) original dirty surface (Sample 7(6)) and (b) fully clean surface (Sample 8(6)).

Table 4 EDX results for the marble samples.

SEM Quant results			Ref: Demonstration data SiLi detector				Spectrum label: Samples 1&2
System resolution = 61 eV			Quantitative method: ZAF (6 iterations)				Analysed all elements
Element	Spectrum type		Element (%)				Compound
			Dirty		Clean		
			Average	SD	Average	SD	
C	K	ED	17.43	2.24	12.70	0.18	$CaCO_3$ 01/12/93
O	K	ED	48.38	2.37	51.27	0.89	Quartz 01/12/93
Na	K	ED	0.24	0.02			Albite 02/12/93
Mg	K	ED	0.74	0.25	0.49	0.05	MgO 01/12/93
Al	K	ED	0.99	0.39	0.11	0.02	Al_2O_3 23/11/93
Si	K	ED	1.89	0.75	0.16	0.03	Quartz 01/12/93
S	K	ED	0.32	0.05			FeS_2 01/12/93
Cl	K	ED	0.26	0.23			KCl 15/02/94
K	K	ED	0.16	0.06			MAD-10 02/12/93
Ca	K	ED	26.65	4.23	35.49	0.97	Wollas 23/11/93
Fe	K	ED	1.57	0.23			Fe 01/12/93
Total			100.00		100.00		

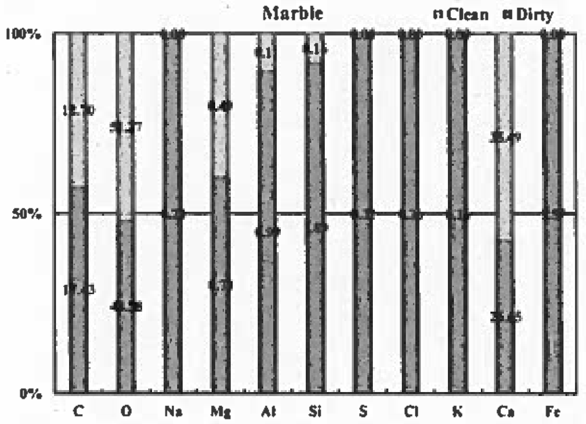


Fig. 17 Chemical elements on the surfaces of the original dirty and fully clean marble samples.

the surface. In addition, since Mg, Al and Si still existed after cleaning, the clean marble likely contained small amounts of Magnesium oxide (MgO), Aluminium oxide (Al_2O_3) and Quartz (SiO_2).

Mineral Zone [31] reported that the main chemical compositions in marble included 3%-30% silica (SiO_2, varying with variety), 1%-3% iron oxide (FeO + Fe_2O_3), 28%-32% lime (CaO), 20%-25% magnesia (MgO) and 20%-45% loss on ignition (LOI). For the marble samples in this study, the detected amounts of silica (SiO_2) and lime (CaO) seemed to be reasonably within the reported range. Iron oxide (FeO + Fe_2O_3) only appeared in the soiling but disappeared on the fully clean surface. The magnesia (MgO) was measured to be much lower than the reported range. The extra chemical elements detected were Na, Al, S, Cl, K and their compounds, but only Al stayed on the fully clean surface and the rest elements disappeared on the fully cleaned surface, which indicates they were part of the soiling.

The test results in this section showed that the chemical substances on the stone and brick surfaces were largely different for different types of stones and bricks. Some chemical elements and compounds largely decreased or increased after cleaning, but the chemical elements C and O always remained at large proportions of all the chemical elements in the stones and brick. The chemical elements and compounds that disappeared may be the main compositions of the soiling deposited on the stone and brick surfaces. As the masonry façade was always exposed to the open environment for a long time and even centuries, chemical reactions would occur, which would nevertheless form various chemical compounds or multi-components on the stone and brick surfaces from the polluting gases in the air.

8. Conclusions

In this study, a series of physical and chemical tests were conducted to extensively investigate the changes in the characteristics of seven different types of popularly used masonry stones and bricks in Edinburgh during the cleaning process, i.e., yellow sandstone, red sandstone, limestone, marble, granite, white clay brick and yellow clay brick. The chemical analysis included micrographing the stone façade and detecting the chemical elements and compounds on the original dirty and fully clean stone and brick surfaces using the combined SEM and EDX techniques. This complete research work has contributed towards the building cleaning in at least three main aspects, i.e. systematic assessment of the physical and chemical characteristics of masonry stones and bricks during building cleaning, detection of the soiling deposited on the surfaces of masonry stones and bricks, and evaluation of cleaning effectiveness using grayscale imaging techniques [22].

The chemical investigations conducted using the SEM and EDX techniques showed that the chemical substances on the stone surface varied largely for different types of stones and bricks. Some chemical elements and compounds largely decreased or increased during the building cleaning, but the chemical elements C and O always remained at large proportions of all the chemical elements in the stones and bricks. As the stone façade was always exposed to the open environment for a long time, chemical reactions would occur, which could form various chemical compounds or multi-components on the stone or brick surface from the polluting gases in the air such as SO_2, H_2S, etc.. This would lead to the formation of the soiling on the stone surface. This study showed the way to detect such soiling using chemical analysis by monitor the changes in chemical elements and compounds during the building cleaning.

In summary, the investigations in this study indicated that the physical and chemical characteristics on the surfaces of masonry stones and bricks were all largely influenced by the building cleaning. For the types of stones and bricks assessed in this programme, a stone or brick with a higher cleaning degree always corresponded to a brighter and harder surface. An appropriate stone cleaning method could not only improve the appearance of the building but also protect the stones from quick decay and damage. However, further protection after building cleaning is still needed. Much effective research work has been done toward this aspect, e.g., using nanocomposites, polymer materials, etc.,as coating layers to protect the cleaned surfaces of historic buildings and monuments from further environmental erosion and weathering [26-29]. Meanwhile, the present study could help to pave the way for selecting more appreciate, economical and effective methods for cleaning existing listed masonry stone buildings. Further research is still under way on these issues and more results will be published later.

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