Lime Binders for the Repair of Historic Buildings: Considerations for CO_2 Abatement

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Abstract

Lime binders are utilised worldwide and are associated with a considerable scale of production and corresponding CO_2 emissions. The relevance of this review is therefore international in scope, with production transcending geographical boundaries and construction practices. An holistic view of lime binders, considering their provenance, production and utilisation offers the potential for significant CO_2 savings urgently required globally. Importantly, the technical aspects of lime materials production is critical, but the alteration of behaviour of both specifier and those undertaking the construction processes is also essential in achieving meaningful CO_2 emission reduction. This review paper investigates the life cycle stages of lime binders in line with the features highlighted in the Scottish Building Alliance (SBA) 'building life cycle stages model'. It attempts to determine what can be learnt from our understanding of the manufacture and use of binders from historic, current and future perspectives in the context of reconciling the production of historically authentic materials in a decarbonising environment. The production and use of such authentic historic lime binders initially appears highly carbon intensive through its utilisation of relatively inefficient kiln technologies and loss of economies of scale associated with larger operations. However, this review shows numerous benefits in the production of such binders; including their CO_2 sequestration capability, lower potential processing energy, and a reduction in excessive 'carbon miles' associated with transportation. Importantly, we show how historic production and on-site manufacture approaches with lime-based materials also offers exciting potential for carbon savings. Importantly, hot mixed lime mortars offer the promise of higher durability materials compared to their modern cold manufactured counterparts, thereby reducing the frequency of repeat maintenance interventions, again yielding CO_2 savings. We suggest hot mixing, whether in an innovative contemporary materials realm or within traditional contexts should be explored. In addition, hot mixed materials can concomitantly satisfy conservation requirements for repairs through better reflecting the aspired to building conservation requirement of like for like materials replacement reflected with historically produced binders. We suggest future paths for the industry that would simultaneously reconcile demands for authentic materials and production methods for the conservation sector with the necessity of achieving cleaner production in a decarbonizing world.

1.0 Introduction

The sensitive repair of historic buildings invariably requires an understanding of indigenous materials that compose their fabric in order to ensure greater levels of compatibility and authenticity upon intervention (Gibbons, 2003; Hughes & Válek, 2003; Clifton-Taylor, 1987). Lime binder technologies play a key role in many building conservation projects given their international prominence in the communication of traditional architecture. Such binders are commonly utilised in a wide range of construction technologies within historic buildings, and include concretes, mortar, plasters, renders, lime washes, and grouts (Forster & Carter, 2011; Bras & Faria, 2017). The scale of their traditional use is today reflected in the cost of repair and maintenance of historic and traditional buildings which is considerable. Indeed, in Scotland alone, ECORYS (2012) estimated that 0.5 million pre 1919 traditional buildings exist ostensibly constructed in lime based materials (5.5 million UK wide), and the estimated spend on repair and maintenance is in the order of £4 Billion in Scotland. Regrettably, as with other binders, lime based materials have been traditionally associated with high environmental impact given the nature of their production. Yet, within today's decarbonizing world it is essential that the sustainable production, manufacture and supply of repair materials are capable of satisfying the societal, economic and environmental demands placed upon them (Brundtland, 1987). Societal demands relate to the retention and support of cultural heritage through compatibility of repair materials, with aspirations to replace on a like for like basis (BS7913; 2013; Bell, 1997; Jokilehto, 1998). Economic demands are contextualised around costs associated with procurement, manufacture, initial and life cycle construction efficiencies, and ultimately deconstruction (Eagan, 1998; Latham, 1994). Environmental demands relate to the aim of producing low carbon materials solutions that attain compatible and high durability fabric repairs (RIBA, 2011; Forster et al., 2015, Forster et al, 2011; Kayan et al, 2016).

Yet the nature of such demands can differ greatly, and reconciling them is extremely challenging, with conservation aspirations to retain and support like for like authentic historical materials difficult to achieve within the environmental, and also the economic, constraints associated with current binder technologies. Although modern methods of lime binder 'production' may be more environmentally and economically efficient in terms of their reduced CO_2 production during calcination, the nature of the lime they produce may not adequately reflect those historically encountered, with current limes lacking critical characteristics such as regional mineralogical specificity. Consequently, the sector is seemingly caught between an aspiration to promote 'traditional' lime binders and their associated technologies on authenticity grounds, whilst struggling to adjust to global austerity economics (Meegan et al., 2014) and increasing environmental regulation and decarbonization (EU emissions trading system (EU ETS), 2018; European Union Paris Agreement, 2018; Stork et al., 2014).

The importance of this cannot be understated, given that the cement industry contributes 5% of global anthropogenic CO_2 (Ishak & Hashim, 2015). Contextualising emissions for the broader construction sector, current

figures for the degree of carbon expended in the UK can be divided into significant stages. Prominently, in contemporary practice the 'before use stages' of construction highlights that; Design expends 1.3 CO_2 (Mt); Manufacture expends 45 CO_2 (Mt); Distribution expends 2.8 CO_2 (Mt); and Operation on site expends 2.6 CO_2 (Mt) (Lawrence, 2015). Attempting to calculate lime's carbon impact, broad frameworks such as the Publically Available Specification (PAS) 2050 (PAS, 2011) attempt to specifically evaluate and establish the main parameters and boundary conditions. Whilst the *'UK Building Black book: capital cost and embodied CO_2 guide'* (Hutchins, 2010) gives more specific CO_2 data on materials, components, and associated construction elements. Furthermore, Hammond and Jones (2008) provide a materials focused inventory of carbon and energy. However, although these sources are recognised as being helpful for objective measurement, they are currently insufficiently nuanced, and fail to include specialised, specific data on lime binders.

Such specific data is key, as lime production in Europe satisfies many applications, with approximately 20% of all lime produced being utilised in construction and civil engineering applications (Stork et al, 2014) and, logically, it proportionately negatively contributes to emissions. Although modern lime production both appears to, and does, offer advantages, here we argue that in fact, additional CO_2 savings can be attained by evaluating historic manufacture and construction practice. This review therefore attempts to determine what can be learnt from understanding the production and use of historic lime binders. The review highlights possibilities that may reconcile the conflicting societal, economic, and environmental demands whilst simultaneously drawing on methods of production and construction satisfying the authenticity required in historic repair. To achieve this, the review evaluates the carbon associated with lime binders on a life cycle basis in line with the features highlighted in the accepted stages of the Sustainable Building Alliance (SBA, 2009) model. Importantly, in order to view lime binders and their potential for CO_2 reduction holistically, the review considers these stages in past (historic), contemporary (current), and potential (future) contexts. For each of these contexts, the review utilises these accepted stages to create a narrative broadly reflecting, the *'before use'* (Provenance, production and construction stages), *'use'* (Building operation performance, maintenance and refurbishment) and *'end of use'* stages (Deconstruction, disposal and recycling) of the materials.

2.0 Lime Binders: 'Before use' stage

In the SBA (2009) model, *'before use'* is subdivided into two major subsets, namely, *'product'*, and *'construction'* stage. Here we present a review of historic, current, and future '*before use'* or production for lime binders.

2.1: Historic lime binders: Production & construction uses

The sourcing of the raw material for lime and its subsequent production are logically connected with naturally available mineralogical deposits (Clifton-Taylor, 1987; Brunskill, 1978; Robertson, 1949). Historically, ease of extraction, lime production technologies, and efficiency of transport were essential for cost effectiveness (Bishop et al., 2017). Over time, broader socioeconomic factors changed in all these areas, reflecting technological innovation relative to labour rates and materials costs ranging from winning the stone to transportation, and man and horse-power (Skinner 1969). Simultaneously, a knowledge of limestone ($CaCO_3$) quality grew, based upon repeated effective empirical use. Nevertheless, despite these changes, many socioeconomic factors remained generally constant over the centuries, with numerous small-scale extraction sites existing in lime bearing regions in Europe being exploited (Bishop et al., 2017; Válek, 2015; Hughes & Válek, 2003).

However, in contrast to commonalities in production and transportation, kiln design varied according to geographical context and resource availability. Common types included 'clamp', 'draw' and later 'horizontal'. Historically, clamp kilns (rudimentary pits dug into the ground) were extensively utilised, and Bishop et al (2017) highlight their key role in UK lime production, noting that in the Braehead area of Lanarkshire alone there were 140 clamp kilns identified. These kilns required considerable skill and knowledge to operate effectively in controlling the burn and ensuring the continuity of product (Bishop et al., 2017). Indeed, such primitive technologies could produce higher quality materials through their slower burning and were often preferred to draw kilns, which tended to 'bring down' the lime before it was thoroughly calcined (Bishop et al. 2017; Nisbet, 2005). From this historical perspective, efficiency in calcination was linked to total conversion of limestone into quicklime (CaO) with a resulting 'soft burnt' product that exhibits rapid reactivity and, importantly, minimised kiln running cost (Válek et al., 2014).

In addition to calcination conditions, it was essential for economic efficiency that kilns were located near to the source of stone rather than the source of coal (Skinner, 1969), given their respective weights and bulk for transportation purposes (Skinner, 1975). Regarding the source of stone, good quality high calcium lime deposits were well known and their continued use was ensured for large-scale deposits, which allowed large-scale extraction and expansion of operations in perpetuity. Conversely, lower quality or smaller-scale deposits were rapidly abandoned or exhausted (Válek et al., 2015), as evidenced by physical topographical traces and historic records

(Bishop et al., 2017; Válek et al., 2015). Industrial development and growing demand for building materials led to new quarries opening in the latter 18th and, more rapidly, in the 19th century (Gibbons, 2003; Skinner, 1969).

Increased materials use led to greater supporting scientific knowledge, in turn informed by better understandings of hydraulic binder production. This was facilitated by the advancing scientific knowledge from the enlightenment (Herman, 2003) and by early chemists and engineers (Smeaton, 1756 (Teutonnico et al., 1994); Black, 1766 (1893); Vicat, 1837) who meaningfully classified and characterised limes (Eckel, 1905; Pasley, 1838). This increased knowledge supported further exploration, and exploitation, of new resources, which were now better understood, thus reducing risks associated with capital intensive entrepreneurial ventures (Skinner, 1969). That said, using impure sources of limestone for burning was not uncommon, and according to individual accounts, the processing of lime to a binder had to be adjusted to reflect its properties (Smith, 2004).

Although advances in production and sourcing knowledge occurred, the transportation of stone remained limited to short distances between kiln locations (Bishop et al., 2017). Kiln type was important to efficiencies here, with flare (flame) kilns requiring as much wood as raw material (i.e. equal in weight), meaning the transportation of stone and wood was volumetrically equal (Válek et al., 2015; Válek et al., 2014). In contrast, draw kilns used coal in a weight ratio of approximately one to six to one to four to limestone, which thus favoured coal as an efficient fuel (Bishop et al., 2017), and coal seams near lime resources were sought. Transportation of stone or fuel over longer distances was only viable if navigable waterways or other cheap transport mechanisms were present (Bailey in Calaria, 1992; NRS, 2018; Bishop et al., 2017). Additionally, burning near the stone source benefited from weight loss during calcination, thus the produced material was approximately 50% lighter to transport (Bishop et al., 2017), even if the volume remained the same (Válek et al., 2015; Válek, 2015). In specific situations for larger building sites (i.e. large towns), an almost permanent supply of lime was required (Válek et al., 2015). In these cases it was also convenient to have limeworks close to, or sometimes even inside, town centres (Marinowitz et al., 2012).

The availability and relative scarcity of fuel was a key factor driving kiln innovation, and a link between the economic efficiency of production was correlated with fuel and haulage costs. Indeed, it took approximately 2 tons of limestone and between 0.33 and 0.5 tons of coal to produce a single ton of quicklime (Bailey, in Calatria, 1993). Given that flame kilns required a 1 to 1 ratio of raw material to wooden fuel in order to produce lime, they came over time to be replaced with coal-fired kilns. Moreover, resource depletion was an issue even in the medieval period, especially regarding deforestation for charcoal production (Wright, 1964), and pollution related complaints about the use of coal in London are noted as early as the 13th to 14th centuries (Salzman 1923; Wright, 1964). Traditional burning and subsequent lime binder processing was extremely labour intensive, with post calcination materials separation based on quality being a fundamental requirement. Consequently, separation as a quality

control mechanism was achieved by rudimentary materials evaluation that compared colour and relative weight to stone volume. As a result of relative burning efficiency, waste was minimal given highly evolved understandings of the stone, the kiln and empirical operative knowledge (Bishop et al., 2017; Válek et al., 2015). In terms of their construction, early kilns had no universally accepted design (Bishop et al 2017) and were operated intermittently, although not infrequently (Bishop et al., 2017). Notably, although CO_2 emissions due to the decomposition process remain the same as with modern kilns, traditional kilns would be considered inefficient by modern standards given their relatively high heat loss and poor control of the burning process. With traditional kilns, the whole kiln needed heating for every burn, and considerable air passed through. Indeed, an historic intermittent flare kiln operates at approximately 20% efficiency (Hughes et al., 2002, Válek et al., 2018) compared to current modern kilns, which operate at approximately 80% efficiency (Holmes & Wingate, 1997).

Table 1
Burning efficiency (calcination process only) of historic kilns

Kiln type	Operation mode	Fuel type	Date	Fuel energy per lime MJ/kg CaO	Fuel Emissions per lime kg CO_2/kg CaO	**Kiln fuel efficiency %**	**CO_2 burning efficiency %**
Open top flare kiln (Válek 2015)	Periodic, small scale, experimental	Wood	Replica of medieval	15.21	1.67	19%	32%
Chamber flare kiln (Válek et al. 2015)	Periodic	Wood	1841	11.67	1.28	25%	38%
Shaft kiln (Hassenfratz 1825)	Periodic / continuous	Charcoal	1825	4.50	0.66	64%	54%
Shaft kiln (Boynton 1981)	Continuous, mixed feed	Coke	1974	4.01	0.32	72%	71%
Shaft kiln (Boynton 1981)	Continuous, annular shaft	Natural gas	1975	4.39	0.25	65%	76%
Rotary kiln (Boynton, 1981)	Continuous, rotary kiln	Oil	1976	7.26	0.56	40%	58%
Rotary kiln (Boynton, 1981)	Continuous, rotary kiln	Natural gas	1977	5.76	0.32	50%	71%

Table 1 outlines the fuel energy requirements associated with lime production. It highlights the energy produced by type of fuel to obtain 1 kg of quicklime, and the fuel emissions and CO_2 emitted by burning fuel. A kiln fuel efficiency is asserted, relating to the ratio of energy required to decompose 1t of $CaCO_3$. The fuel energy per lime CO_2 burning efficiency is the proportion of CO_2 emitted by limestone decomposition ($CaCO_3$) over the total CO_2 emissions (fuel burning and limestone decomposition). Table 1 shows that fuel efficiency of historic kilns was poor, which is in contrast to today's kilns that can operate with fuel efficiency above 95%. The increasing fuel efficiency subsequently increases the proportion of CO_2 emissions from the process of limestone decomposition itself. The proportion of CO_2 emitted by limestone decomposition ($CaCO_3$) over the total CO_2 emissions of such highly economically efficient kilns can be approximately 80% depending on fuel type. The elimination of heat loss and the quality of fuel used are the main parameters to control CO_2 emissions today, whereas, historically, efficiencies

driven by fuel costs only indirectly affected CO_2 burning efficiency. In the past, kiln efficiency and conversion of $CaCO_3$ into CaO would be critical for the economic viability of the operation, with repeat sales of lime being based upon cost, quality and production volume. Notably, it was quicklime that was historically the final product manufactured and sold to consumers (Lynch, 2007; Lynch, 1998a & b), a factor that differs radically from modern lime production with today's prevalence of the sale of calcium hydroxide [$Ca(OH)_2$] based products. Indeed, this had significant implications historically for handling and utilising materials given quicklime's inherent reactivity. The transportation and storage of quicklime, even though it was highly reactive in the presence of water, was possible as the lime was in lump form (Smith, 2004) and therefore had a relatively low surface area to react with moisture. Nevertheless, great care in protecting burnt lump lime from direct contact with water was particularly important when transporting quicklime in loose form on carts or via waterways sealed in barrels (Bailey, in Calatria, 1993).

The transportation and storing stages of quicklime for construction had different requirements to modern hydrated powdered materials and played a much more significant role than is generally noted in contemporary literature. Practically, a link between the mineral composition of burnt lime and its reactivity was an important characteristic; high calcium limes were extremely reactive, whilst eminently hydraulic quicklime exhibited only slow reactivity as a function of increasing hydraulicity (Smith, 2004). Reflecting this, historically, various methods for storage were employed, enabling combined storage of lime simultaneously with the slaking and mixing processes. Common methods (Forster, 2004; Holmes, 1993; Válek & Matas, 2012; Jedrzejewska, 1967) across Europe included mixing and covering wetted lump lime with a layer of sand (i.e. sand slaking and hot mixing) (Lynch, 1998 a & b; Forster, 2004; Holmes, 1993; Válek & Matas, 2012; Jedrzejewska, 1967; Moropoulou et al, 1996; Margalha et al., 2011; Smith, 2004). This lime could be left for a reasonable length of time, limited further slaking, and obviated carbonation of the lime that had converted to $Ca(OH)_2$ (Smith, 2004). Products of a lower quality were not wasted, but construction processes were adapted to reflect the practical restrictions of utilising greater variability in the binders (Válek et al., 2015). In the subsequent production stage, transforming burnt lump lime into a functional binder had many potential permutations (Smith, 2004). Notably, from an efficiency perspective, these materials potentially had direct energy efficiencies compared with today's site practice. One key benefit in the use of hot mixed mortars is the obviation of CO_2 / energy input associated with milling (see Ishak & Hashim, 2015) of materials, which can be considerable.

The scale of the building project was also significant in the context of transportation and storage of quicklime. The supply of lime to large projects (i.e. cathedrals and infrastructural masonry structures) was clearly more logistically complicated, requiring greater capacity for storing and processing quicklime (Smith, 2004). Stockpiling was essential to maintain construction productivity, especially for materials needing to be transported great distances. The specific binder type (non-hydraulic or hydraulic) influenced storage approaches and considerations for site

establishment. Lynch (1998a) notes the impracticality of storing non-hydraulic lime for bedding mortar as the amount required would have required huge storage pits for the lime needed, making it an unrealistic proposition (e.g. 20 19th century workers laying a thousand bricks a day each would require approximately 20 tonnes of lime a day). Also, non-hydraulic quicklimes had to be protected from CO_2 ingress, something traditionally achieved by slaking them to a putty and using excess water as a seal, which also improved the functional properties via maturation (Margalha et al., 2013).

In addition to the slaking process of free lime causing the disintegration of lumps into a powder, certain compounds react in presence of moisture (hydrate), creating stable bonds and forming the skeletal matrix structure of the material. The hydraulic reaction speed depends on the nature of these hydraulic compounds, but is subject to the ambient temperature, relative humidity, and quantity of liquid water used to dampen the material (Barnes; 1983; Hewlett, 2003; Taylor, 1997). Therefore, a mortar produced adopting the mixing of un-slaked natural hydraulic lime with sand could benefit from the hydration of hydraulic compounds present in the binder, creating an intimate, often complex relationship between aggregate and matrix (Forster, 2004). Importantly, mortars made with un-slaked and screened non-hydraulic lump lime inevitably contained particles related to the original composition of raw limestone material and burning technology, thereby increasing its inherent complexity (Válek, et al., 2015).

Compositionally, historic mortars are generally associated with higher binder to aggregate ratios than their modern counterparts. It was not uncommon to see ratios of 1:1 or 1:1½ (Smith, 2004; Lynch, 1998a; Lynch, 1998b; Lynch, 2007). This partially compensated for deficiencies in preparation processes and helped ensure durability and strength (Moropoulou et al, 1996; Margalha et al, 2011). In terms of energy expenditure and CO_2 emissions, these ratios were clearly inefficient; however, this must be considered alongside the lower process energy associated with hot mixing. Historic processing methods for lime logically related to the construction purpose and installation process the lime was intended to be used for. Each construction process type would have minimum requirements for the degree of slaking directly linked to the nature of the quicklime (Smith, 2004). For example, bedding mortars may have been rapidly used even if *insitu* slaking was ongoing during the building process, whereas this would be impossible for stuccos, as popping and pitting would occur, detrimentally affecting the finish.

2.2 Current lime binders: Production & Construction uses

The historic economic efficiencies espoused by Skinner (1969) in transporting fuel to the stone arguably reflect current ones. Logically, both historical and modern kilns are located as close as possible to the quarries to increase economic efficiency in transporting limestone. However, the total number of current large-scale kilns is significantly lower compared to historic, small scale burning kilns (Skinner, 1969; Bishop, et al., 2017; Válek et al., 2015). Moreover, sourcing of limestone and its calcination has undergone significant rationalisation. Many current lime

quarries producing binders are increasingly large 'super quarries', with a much larger scale of operations. Extraction processes are now well understood, and quarries benefit from advanced mechanised technologies in almost all areas of production. A better understanding of the nature of limestone, and its homogeneity and variability according to deposition and strata (Boynton 1981, Oates 1998) also underpins current practices. Indeed, accurate mineralogical identification determines current processing functions and end use; for example, whether specific quality means the limestone should be utilised for cement or lime manufacture, or simply for gravel and aggregates in alternative construction purposes.

Generally, cement production has made significant advances in kiln efficiency and pollution abatement (Vatopoulos & Tzimas, 2012; Benhelal et al, 2013), but this has not been as pronounced in production in limes (Livesey, 2007). Undoubtedly, a critical factor related to CO_2 levels in current production of cementitious materials is associated with the fuel used (whether coal, coke, oil, gas, wood or biomass) during calcination (see Table 1). The nature of the fuel influences CO_2 output, and, importantly, calorific value and its reduction has been the focus of both legislation and technological innovation. Large scale, multi-kiln plants are common and can potentially utilise different fuel types, thereby reducing fuel costs through capitalising on economic market fluctuations (purchase or substitution). More recently, introducing waste products or biomass (Ishak & Hashim, 2015) as fuel has created further environmental savings. Yet, despite recent analysis (Stork et al., 2014) highlighting CO_2 related efficiencies through the use of biomass, its uptake has been hindered due to its higher economic production costs (Ishak & Hashim, 2015).

Although advances in kiln technologies have occurred (Livesey, 2007), innovative kiln design remains key to emission reduction efficiencies. Indeed, Livesey (2010) asserts that different types of kiln are in operation in the production of lime, but their energy efficiency is variable. Objectifying the energy, comparative data ranges from 2,500 kcal/kg for a basic batch kiln to 750 kcal/kg for a particularly efficient shaft kiln (Wingate, 1985, cited in Livesey, 2007). Livesey (2007) notes a value of 800 kcal/kg as being a reasonable estimate of performance, which is comparable to a general cement kiln with higher throughput, and thus economy of scale. Indeed, Stork et al., (2014) emphasise that the greatest opportunities for efficiencies are in replacing horizontal kilns with vertical kilns, leading to abatement costs of approximately 38 euros a tonne (at 2014 prices see Stork et al., 2014).

Other approaches to reducing CO_2 produced in the calcination process are associated with Carbon Capture and Storage, or CCS (Ishak & Hashim, 2015; Sepehri & Sarrafzadeh, 2018), which has received much interest despite being in its infancy. However, Hill et al (2016) observe that the capture dimension of CCS is both expensive and energy intensive, with Ishak & Hashim (2015) indicating that the capital investment of retrospectively installing CCS to existing plant is largely cost prohibitive, hindering uptake.

Considerable energy is associated with the transportation of lime (Crishna et al., 2011), especially as in current practices, most lime is associated with internationally produced lime binders from France (St Astier, 2019), Germany (Otterbein, 2019), or Portugal (Cimpor, 2019) that are exported worldwide (incl. UK and USA). Importation of these materials arguably hinders effective implementation of green procurement strategies (APRES, 2018) via increased carbon miles. Indeed, most kilns are remote, largely to obviate issues of community tensions by putting them at a distance from population centres (Pyke & Stummer, 2003), but this logically results in greater transportation distances for CaO (if unconverted). The subsequent conversion of CaO into $Ca(OH)_2$ also requires operational energy and capital investment in specialised equipment to crush and mill the lumps of quicklime, store the ground quicklime, hydrate the lime with water, separate large pieces, and deliver the crude hydrate to the silo (Brown 1996). Logically, all these stages require operational energy and additional processing capacity. The increased optimisation of slaking plants are arguably more pronounced in current materials due to aspirations to attain a more uniform particle size and general uniformity of product demanded by specifiers.

However, a growing recognition of utilising CaO for 'hot mixing' of lime mortars is noted (Forster, 2004). These arguably yield more authentic limes used in historic construction, and, significantly from an environmental perspective, obviate energy intensive conversion stages. Thus, this is one area of considerable sector interest (BLF Hot-lime forums Ireland, 2018; Technical University Dresden, 2018) in achieving authentic production and at the same time attaining CO_2 savings (Forster et al., 2004). Far greater environmental efficiencies and materials authenticity in lime production could be achieved with higher levels of indigenous lime production in importing countries. This would simultaneously reduce the carbon 'miles' associated with product to market and potentially create materials better reflecting those historically used. Significantly, from an authenticity perspective, current materials differ in many ways from historic hot lime mortars. Today, production of hot lime mortars is generally considered a 'fringe' or 'niche' sector activity, although growing interest has been noted (BLF Hot-lime forums Ireland, 2018; Technical University Dresden, 2018) in re-adopting these materials and techniques from the analysis of historic mortars and historic literature based documentary evidence. Anecdotally, they are considered more durable (Moropoulou et al, 1996; BLF Hot-lime forums Ireland, 2018), with reduced materials costs and a notable ease of use that increases construction productivity (Forster 2004; Válek & Matas, 2012).

The consistency and refinement of mainstream anhydrous products reflects greater advancement in burning technologies and higher levels of scientific knowledge of multi-phase compounds. Technical literature supporting lime binder manufacture has also advanced, with products now being more consistent and supported by both EU norms and British standards (BS1015, 1999; BS459, 2015). These standards underpin current manufacture technologies, with tests logically following production. In addition, testing materials in 'real time' during calcination and slaking of their hydrated production has enhanced robust quality control mechanisms, thereby assuring product

conformity. However, the same cannot be said about hot limes, with research in its relative infancy (Moropoulou et al, 1996; Válek & Matas, 2012). Further, rigorous expectations in performance and an increasingly litigious society reinforces the need for a uniform product (Strike, 1991) and an increasing prevalence of the use of premixed lime products is also noted (Torney & Forster; 2012). Premix formulated materials are sold on the virtue of relative ease of use (Torney et al., 2015), with little expert craft or specific knowledge. Higher hydraulicity binders are also currently more prevalently specified and used given their greater likeness to cements (both powders), ensuring operative familiarity with process (Forster & Carter, 2011; Forster et al 2014; Banfill et al, 2016).

In contrast with historic materials mortar manufacture, most current materials are produced adopting dry bagged hydrated $Ca(OH)_2$. They are batched on site, adopting forced action pan or belle mixing methods (Gibbons, 2003). More recently, the increasing deployment of silos (Limetech, 2018) has helped control proportions, compositional distribution, mix times, and water ratios. Yet, the nominal batch ratios commonly utilised for mortars is 1:2½ - 1:3 by volume and this therefore represents a departure from traditionally high historical binder to aggregate ratios (i.e. 1:1½) (Gibbons, 2003; Forster, 2004), and thus from the production of historic mix proportions. Indeed, batch proportions are important for the carbon footprint of mortars, and as Livesey (2010:18) indicates, ratios vary from "1:6 for a Portland cement (PC): sand aerated mortar to 1:2 for an NHL2: sand (1:3 for NHL 3.5: sand and 1:4 for NHL5: sand). This results in carbon dioxide emissions of 175 t/m^3 for PC: sand for NHL2: sand, 210 t/m^3 for NHL 3.5: sand and 170 t/m3 for NHL 5: sand" and clearly highlights the relative range of emissions associated with specification and design choices.

Notably, from an environmental efficiency perspective, one relatively neglected area of investigation arguably key to efficiencies is broader evaluation of the embodied carbon associated with lime binders' carbon sequestration. Indeed, Figueiredo, Ball and Lawrence (2016) note that as a concept, sequestration is acknowledged as being important in Life Cycle Assessment (LCA), but is not included in any primary figures in inventories of carbon and energy. Clearly, the review presented here shows how such binders can re-capture any CO_2 released during calcination. What is more, Livesey (2010:18) highlights that calculating the carbon footprint of production involves conversion of "the measure of efficiency to carbon dioxide emitted per kilogram of product." Importantly, for hydraulic limes the calcium carbonate requirements are much lower than for rich air limes, as Livesey (2010:18) notes: "the CL90 could be estimated as 0.88 Kg CO_2/kg compared with around 0.77 kg CO2/Kg for natural hydraulic lime and Portland cement". Indeed, Hill et al (2016) indicated that the majority of CO_2 produced in cement manufacture is associated with the dissociation of CO_2 from the limestone. Pure high calcium lime is, in theoretical terms, in CO_2 equilibrium; with the carbon released during burning being reintroduced during the setting process. Thus, CO_2 is chemically rebonded during the carbonation setting process (Allen et al., 2003; Lawrence & Walker, 2008; Lawrence et al., 2007). Here therefore, such emulation of historic limes by production of high calcium lime

binder can be considered carbon neutral in limestone disassociation terms if sufficient time for full carbonation is given.

Nevertheless, the situation is not without its complications. For example, practicalities, such as an inability for CO_2 to diffuse through to the core of mass wall construction (Tuetonico et al., 1993) can mitigate against this, and some lime even takes decades to fully convert (Forster et al, 2014). Also, certain materials providing hydraulic qualities (e.g. pozzolans) can react with lime, forming compounds that limit CO_2 absorption even further (Alberti, 1992; Vitruvius, 1914). Similarly, the burning of impure limestones and carbonate rocks results in the formation of calcium silicate, aluminate and ferrite compounds, and phases that hydrate and create hydraulic compounds, but other non-reactive calcium phases are also noted. Therefore, natural hydraulic limes, when compared to current limes, cannot be said to recapture all the CO_2 released during calcination, and the higher their hydraulicity, the lower is the proportion of available free lime to carbonate (Forster & Carter, 2011; Forster et al, 2014, Banfill et al, 2016). Despite this, they offer clear environmental efficiencies through the obviation of CO_2 released during milling, and, at the same time, their potential for authenticity in the repair of historic buildings is far higher than the consistent hydraulic binders produced today.

2.3 Future lime binders: Production & Construction

Economic assessment and analysis by large companies has become increasingly sophisticated, with more data being theoretically available. Today, quarries face tougher and more complex environmental and legislative controls on production regarding the environmental impact of extraction and its effects on local communities (Pyke & Stummer, 2003). In the future, it can safely be assumed there will be a greater recognition of the relative scarcity, and geologically finite nature, of high calcium limes. This is particularly so given that purity is not necessarily required for most building purposes, and that high calcium limes could be better utilised for chemical (Pharmaceutical etc.) or other industries (Steel making etc.).

Regarding technologies for future production, current kiln design has now reached almost optimal thermodynamic efficiency with corresponding carbon savings through the obviation of heat loss. In addition, notable efficiencies are associated with the fuel utilised to calcine lime and arguably, future practices will see a greater variety of recycled waste product materials continually increase in use alongside bio mass fuels. Importantly, it is recognised that future emissions reductions may benefit from Carbon Capture and Storage (CCS), Carbon Capture and Utilization (CCU) and kiln flue scrubbing technologies at source (Stork et al., 2014; Ishak and Hashim, 2015). One logical mechanism to reduce future fuel emissions is to reduce firing temperatures, but such an approach is not without its challenges. Firstly, this requires a comprehensive understanding of both the long-term performance and applicability of low-fired binders (Hewlett, 2003; Taylor, 1997). Secondly, it requires knowledge of the materials

produced relative to those historically encountered. Importantly issues such as the thermodynamic stability and durability of the products of hydration formed at lower kiln temperatures, and, more specifically, the α, β, γ polymorphs of Belite (C_2S) are essential (Taylor, 1997; Bonen et al., 1994).

Regarding future materials, recent advances in geo-polymers and alkali-activated binders utilising natural pozzolans or silicate by-product materials show potential as substitutes (Provis & Van Deventer, 2009). Natural pozzolans formed from volcanic activity offer potential for significant change in the sector in their ability to form stable products of hydration without carbonate calcination (Provis & Van Deventer, 2009). However, volcanic deposition and therefore their continued supply cannot be guaranteed. Moreover, these alternative binders may find limited uptake in conservation and restoration projects due to their matrix compositional differences with historic binders, and could be argued to be alien to those noted in the building fabric.

Arguments for a greater future utilisation of indigenous binders is underpinned by their ability to attain greater authenticity in repair mortars for historic buildings within the locale. This can be seen as a return to historic selection of materials, encapsulated in the *'one-mile rule'* of sourcing, whereby most materials for construction of vernacular structures were commonly procured within a notional 1 mile radius of the site, with clear benefits in reducing carbon miles in transportation and haulage costs. Regional lime production is therefore philosophically more defensible when evaluated on a like for like materials replacement basis (BS7913, 2013; Forster et al. 2018; Forster, 2010a & b), and indigenous materials on the whole better facilitate this. These materials have supplementary economic benefits in so much as they offer the potential to stimulate local employment. It must however, be qualified that whilst the materials are important, process is also critical, with hot mixing methods gaining traction relative to cold manufactured mortars adopting $Ca(OH)_2$. These materials better reflect historic materials encountered, enhance performance, and are growing in popularity (BLF Hot-lime forums Ireland, 2018; Technical University Dresden, 2018)

For the future, it is thus arguable that the lime production sector is at a significant juncture; one that aspires to reduce financial and carbon cost, attain higher longevity, and at the same time enhance authenticity in repair mortars. Such aspirations may at first sight appear diametrically opposed, with small-scale regional production adopting indigenous lime resources achieving higher levels of authenticity, but greater costs in manufacture due partly to an absence of economies of scale and small scale kiln inefficiencies. Yet, as this review shows, they nevertheless offer advantages in lower CO_2 associated with transportation of lime feed stocks that may offset any of the aforementioned small-scale kiln inefficiencies. Indeed, 'Green' procurement strategies and increasing interest in 'Green' supply chains (APRES, 2018) show potential to reduce carbon emissions, and lime binders are no exception. Logically, much commonality exists with modern day lime production, but the increasing carbon

reduction narrative adds complexity to the real costs associated with production. For example, a carbon tax must now be considered (EU Emissions Trading System, 2018) and can be particularly high for lime production given its high carbon intensity (Stork et al., 2014). Clearly, binders that utilise lower energy inputs during manufacture are sought, reframing the drivers for innovation historically associated with the financial costs of labour, raw materials, and haulage. This reframes dialogue and creates a narrative for environmental analysis towards materials sourcing, substantiating the credibility of 'Green' procurement strategies (APRES, 2018), with reduced transportation of raw materials being key (Crishna et al., 2011).

Another key consideration within the context of CO_2 reduction, is that it can be assumed that lime binders will be increasingly utilised and deployed in future Modern Methods of Construction (MMC) technology applications (Nadim & Goulding, 2010). For example, MMC currently constitutes 15-20% of construction in the UK, having grown rapidly from 2% in 2003 (Buildoffsite, 2018), and growth is likely to continue. Lime may facilitate this, enabling a move away from energy intensive binder technologies (i.e. Portland Cement) to help facilitate the UK's aspired 33% emissions reduction. Given this, future adoption of lime in construction may be arguably confronted with a twin model; one associated with MMC applications that almost completely eradicates operative skills via factory controlled environments and the other, that retains and enhances the knowledge and advanced skill levels required for bespoke conservation repair projects. Arguably, these latter skills will become increasingly decoupled from the procurement and delivery of new build applications. The future will arguably see traditional trade skills and the high educational investment required for them (Torney & Forster, 2012) undergo a notable reduction within a context of MMC (volume, panel and hybrid) systems uptake (Pan et al., 2007; Ross et al., 2006). Indeed, recently, construction adopting lime binders has been associated with the progressive ease of use ostensibly responding to trade skill deterioration. This has manifested itself in the increased use of modified lime binder and mortar technologies and (Torney et al. 2014; Torney, et al., 2015; Torney & Forster, 2012) (akin to OPC binders) has naturally repositioned the sector on a trajectory away from replacing lime based materials on a like for like basis.

However, this has not necessarily yielded better materials performance, and has arguably reduced the authenticity of repair materials. Importantly, although the conservation sector aspires to better repair of historic structures, it is hindered by training and labour cost efficiencies driven by the main construction sector. Indeed, lime binders currently available benefit from economies of scale in production that reduce consumer costs. These materials, by virtue of mass production, have high quality control and conform to standards developed by the main construction sectors (BS459, 2015; Allen et al., 2003). These standards facilitate performance and quality control in production during construction through, for example, measurable water ratios and silo technologies that obviate water and batch ratio inconsistencies. Other benefits of less complex binders are their relative ease of use and uniformity, enabling conformity to modern specification standards and regulations. Within this context, the litigious nature of

construction and materials specification clauses will continue to drive increased uptake of formulated and pre-mixed hydrated bagged lime and lime mortar products. Here then, this specific future direction would see the standardisation and further focus on efficiencies and qualities of binders take precedence over any production of binders that emulate historic and authentic practices.

Yet, at the same time, as the above review shows, another trajectory the industry could simultaneously follow is one that reintroduces the manufacturing practices and processes once commonly used in the sector in the form of hot mixing. Not only would this help emulate historic production methods and thereby produce lime binders more defensible from a philosophical perspective that are arguably closer in authenticity (Forster, 2010a & b; Forster et al, 2018) to historically produced binders, but also ones with possible environmental efficiencies. In local production this would reduce the need for many stages, thereby helping obviate CO_2 production, and would also lead to greater carbon recapture over time. Further, from an authenticity perspective, currently produced less complex binders do not resemble inherent variations associated with existing historic materials and thus cannot achieve like for like materials replacement (Figueiredo, 2016). The repair and conservation sector will in future arguably require alternative educational strategies to support the retention of traditional craft skills essential for the large proportion of existing traditional building stock (NHTG, 2018). Yet, it is arguable that adoption of this trajectory alongside current directions will help reconcile the production of authentic materials with environmental efficiencies, and therefore holds significant potential for the future.

3.0 Discussion: Past, present and future efficiencies

Local small-scale production and networks have now been almost completely supplanted by large-scale production, with extensive international distribution networks that are ostensibly on a different order of magnitude. The burning of lime has gone through significant enhancement over time, most specifically noted in current kiln design efficiencies and through fuel substitution. Although this has helped attain almost thermodynamic minimum energy in kilns, it means additional carbon savings efficiencies are limited in this context. Significant sector interest is noted in Carbon Capture and Storage (CCS), which offers future potential to obviate large scale emissions in production, and its development is a key aspiration of the industry (Hill et al, 2016; Benhelal et al, 2013).

Yet, in the context of authenticity and producing like for like materials, this path of development has impacted on, and if followed further would continue to impact on, appropriate conservation and repair materials for historic buildings. Indeed, regional bespoke indigenous mortar technologies resembling the complexity of those historically used are in general no longer currently produced. This is despite recent advances in the understanding and characterising of historic mortars (Radonjic et al, 2001; Válek et al, 2012). Somewhat paradoxically therefore, today, we better understand what characterise historic mortars, but have a reduced ability to achieve like for like

material replacement. Ultimately, whilst current internationally produced lime based materials offer a relatively efficient substitution potential over Portland Cement on a technical performance (Rodrigues & Grossi, 2007) and environmental basis, they do not reflect the materials historically encountered, thereby hindering our ability to sensitively conserve historic buildings. Indeed, today's conservation sector appears caught between the aspiration to recreate lime based materials from small scale manufactured regional binders, and the pragmatic allure of standardised mass produced limes that facilitate conformity to robust specification (BS459, 2015), design and performance (BCIS, 2009), and are at first sight far cleaner from a 'before use' or 'production' perspective. Importantly, their uptake is an understandable and logical response to avoid construction claims and wider project litigation that are common place.

Thus, the current developmental path that has been followed means that the production and procurement of lime binders in quicklime form has been largely superseded by slaked hydraulic binders. However, this substitution is, perhaps surprisingly, relatively recent, and mounting archaeological and archival evidence shows that hot mixed mortars were widely adopted and present in British Standards even as late as 1951 (BSI, 1951). Furthermore, at first sight the path that has been followed would suggest that in terms of cleaner technology the last thing the industry should do is to reintroduce regionalised small-scale production of lime, given the kiln inefficiencies. There are however, key potential benefits of small-scale regional production and the reintroduction of historical processes within a contemporary architectural and construction realm. Indeed, there are significant gains to be made in CO_2 abatement from a transportation perspective with regional production given the huge reductions in distances from source of production to site (carbon miles). There are also significant gains in CO_2 abatement to be made from production of non-hydraulic and hydraulic hot limes given the obviation of the latter stages of production. In terms of sequestration, traditional lime binders have a far higher level of sequestration than their contemporary and less authentic counterparts. Finally, anecdotal evidence suggests (Moropoulou et al, 1996) that the durability of hot limes means they last far longer than currently produced binders. Within a repair and maintenance context, small cumulative savings associated with these materials translate into meaningful carbon gains over the life cycle of the building (Forster et al, 2011).

In terms of continuing to conserve historic buildings, there would also appear to be a number of benefits to reintroducing such production alongside modern approaches. Whilst never truly being capable of philosophically matching historic binders, reintroducing past processes would undoubtedly involve the use of indigenous materials. Matching kiln manufacture design and conditions, yielding binders of similar mineral composition and adopting aggregates of comparable geological type and grading will simultaneously go some way to attain materials historically encountered, greatly enhancing authenticity and also moving us towards satisfying current goals for cleaner technology and materials performance.

4.0 Conclusion: gaining environmental efficiencies and historic authenticity

Lime binders are used internationally and are considerable in their scale of production. As this review shows, viewing the provenance, production and utilization of lime binders holistically yields simultaneous possibilities to achieve greater levels of authenticity in historic materials for traditional building repair, and lower environmental impact. Indeed, despite common perceptions that historic manufacture techniques supporting lime use are CO_2 intensive and should be minimised, they may actually offer significant potential for urgently needed CO_2 abatement, whilst enhancing authenticity of repair. To date, such potential has not been meaningfully realised.

Regarding production, kiln design advances have progressively helped achieve almost thermodynamic optimal efficiency. A move away from coal and the adoption of a broad range of fuel substitutes is gaining traction, resulting in meaningful CO_2 reductions. More recently, important advances in CCS have been made, offering the promise of cleaner production with potentially radical implications for the sector. Yet, such developments, whilst creating much needed CO_2 reductions, rarely produce materials close to those historically encountered. Reflecting this, there is now growing sector interest in the reintroduction of use of traditional hot mixed lime mortars, particularly when produced and used locally as was characteristic in the past. The inherent ease of use of such mortars, their ability to sequester carbon during setting and hardening, and their simultaneous enhancement of durability (Moropoulou et al, 1996) arguably justify their wide scale reintroduction into construction practice. Importantly, there are many potential benefits in their reuse for environmental and economic efficiencies, and the simultaneous advantage of attaining greater authenticity in fabric repair.

Nevertheless, large-scale production may not be possible, and the skills required in their deployment may require significant training in a sector that is moving away from traditional skills towards MMC. That said, the utilisation of hot lime mortar techniques need not be a binary choice, with potential for historic hot lime manufacture processes sitting alongside innovative modified lime based materials within an MMC capacity. This dual approach, coupled with developments in kiln technology advancements and CCS, may enable the environmental tensions to be minimised in the area of production of lime-based mortars for both new-build and also historic buildings. However, such substitution of binder technologies or increased lime materials uptake can only be achieved through both effective education for specifiers, contractors and clients, and by robust data collection relating to the holistic performance of the products and their interaction with host materials and architectural technologies. In this way, both confidence in lime binders will be engendered, and, importantly, a broader recognition of their positive benefits in facilitating traditional and historic building performance as historically intended should be achieved. Ultimately, such changes aim to support continued fabric longevity, and help reorient views on the appropriateness of fabric repair. Within this context, logically, small interventions undertaken over a building's life cycle adopting greener materials solutions can cumulatively amount to significant long-term CO_2 savings. The adoption of regional

materials, their manufacture processes and application theoretically reduce the embodied carbon of the intervention, and can help attain highly durable repairs that achieve the aspired like for like fabric interventions with inherent greater authenticity. Conversely, the increased utilization of homogeneous contemporary binders sourced from geographically distant locations limits our abilities to appropriately repair our historic structures in terms of materials, and simultaneously, may fail to benefit from the degree of durability and long term CO_2 reductions attainable through the use of historic binders.

Acknowledgements

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