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Abstract

The effect on porosity (including absorption and sorpitivity) of cement paste and mortar/concrete, of limestone addition to Portland cement is assessed. Based on globally sourced literature published in English since 1993, consisting of 171 publications from 35 countries. The data analysed were from wide ranging tests. The effect on pore structure was also examined in terms of type of Portland cement and limestone, cement fineness and method of producing it, curing, maturity and water-cement ratio, as well as the cement composites with fly ash, slag (GGBS), silica fume and metakaolin and related to strength. Overall, it is suggested that though the use of limestone up to 25% with Portland cement should not impair the pore structure, limit on limestone content for its effect on strength is likely to be about 15%. This should be considered where higher proportion of limestone content is allowed in the Standards.

Keywords: Limestone addition, Portland cement, blended cements, pore structure, concrete, strength.

1. INRODUCTION

Although limestone has been one of the base materials used in the manufacturing of Portland cement (PC) ever since it was first developed in the 1824, its use as an addition to cement is relatively new and is closely associated with the global sustainability drive. In fact, it is claimed that the use of limestone can lessen carbon footprint of concrete by about 15% **[147]**, as well reducing the demand for primary raw materials for cement by around 10% **[130]**. Furthermore, limestone is also a cheap material and easier to handle for cement manufacturers; it results in lowering grinding effort **[15]** and it is available at the cement plant**.**

The use of limestone in the form of blended cement in combination with PC, now commonly known as Portland limestone cement (PLC) was first attempted in 1965 in Germany. French Standards adopted its use in 1979 and in 1983 the Canadian Standards permitted 5% limestone to be combined with cement **[70]**. In 1992 British Standards allowed the use of limestone up to 20% and now its use is recognised in the national Standards worldwide **(Table 1)**, with the maximum limit for limestone content in some cases increased to 35% (BS EN 197-1:2011 [22], EN 197-1:2011 [52], SANS 50197-1:2013 [140], SS EN 197-1:2014 [137] and NMX-C-414-2010 [105]).

As to be expected, the usage of PLC has been recently increased rapidly, particularly in Europe **[79]** and a great deal of research has been undertaken and published on its performance relating to the fresh and hardened states of concrete, as well as the various aspects of durability. However, the published literature was remained fragmented and this does not help in assessing the sustainable use of the material, in a realistic and reliable manner.

A long term view of promoting sustainable construction, calls in question how the use of PLC may affect the permeation and durability of concrete as well in fundamental terms the development of its pore structure which is commonly referred to in practice as porosity and commonly measured as absorption and sorpitivity. Whilst the importance of porosity on the durability of concrete is widely recognised, the relationship between the two is not so clear particularly with different cement type.

Given the above, the study has been initiated to examine the global literature, published in English by analysing and evaluating the published data to assess the effect of limestone addition on the porosity of concrete.

2. AIM AND OBJECTIVES

The main aim of this study is to assess the effect of limestone use as cement component on the porosity, (including water absorption and sorpitivity) of cementitious mixtures (such as cement paste, mortar and concrete) and establish how this may be used in practice. In achieving this aim, the following objectives have been adopted:

(i) To develop and overview the published literature on the effect of PLC on porosity, and for practical reasons, including absorption and sorpitivity, of cementitious mixtures in the form of cement paste, mortar and concrete.

(ii) To analyse and evaluate the published data on porosity (including absorption and sorpitivity) of concrete in a manner that may suggest an optimum level for limestone addition use in concrete.

(iii) To propose the ways for the use of PLC more efficiently in practice.

3. THE EFFECT OF LIMESTONE ADDITION

Overview of the observed effect on the pore structure, in terms of porosity, water absorption and sorpitivity, of cementitious mixtures (paste, mortar and concrete) of using limestone addition, similar to the specifications adopted in the Standards such as **BS EN 197-1:2011 [22]**, is presented in **Table 2**. This is based on the preliminary study of the data presented in the literature, consisting of 97 studies published over a period of 23 years from 1993 to 2015.

Given the large number of parameters involved across the literature, the only viable option available was to initially examine all the data with limestone use relative to the corresponding PC mixtures and assign them to one of the five categories w.r.t. porosity and related properties, expressed as being:

(i) Higher than PC.
(ii) Lower than PC.
(iii) No change.
(iv) Variable, where the relative figures change with limestone content
(v) Unclear, where reference PC data has not been provided.

Additionally, the causes for the observed data as stated by the researchers in each case have also been summarized in **Table 2**. The physical and chemical effects are mainly identified as the filler, heterogeneous nucleation and dilution effects **[77]** as follows:

- The filler effect of limestone refines and improves the porosity of the mix and in general terms results in lowering the water demand for a given workability **[77]**.

- Heterogeneous nucleation takes place since limestone particles work as nucleation sites, raising the early hydration of cement and, consequently, creating an additional mixed up crystallization of calcium silicate hydrate **[135]**.

- The dilution effect acts in reverse to the filler effect and heterogeneous nucleation. The dilution effect is an outcome of the reduced cement content and, as a result, an increase in the effective w/c of the test mix **[77]**.

The above effects are in essence considered to rely on the amount and fineness of limestone addition used in a particular mix **[135]**.

The obvious overall impression to emerge from the preliminary initial examination of the literature as presented in **Table 2** is one of split opinion regarding the effect on porosity and related properties of cementitious mixtures (cement paste, mortar and concrete) with limestone use as cement component.

Further examination of the literature revealed that the variability observed in the initial appraisal of literature appears to be caused by the number of parameters involved in the reported studies and adding to the assessed variability of the effect of limestone inclusion, such as the:

(a) Proportion of limestone used in relation to Portland cement content ranging from 2 to 50% and about one-third of the studies used only one limestone proportion level.
(b) The different test methods used and the procedures adopted, the age at test varying from 1 to 365 days and with it the limestone effect not remaining constant throughout.
(c) Water/cement ratio varying from 0.35 to 0.79 and with a given mix the PLC effect varying.
(d) Fineness of cement mixture and mineralogical composition of Portland cement.

Notwithstanding the above, and the uncertainties thus arising for not being able to establish a clear consensus on the effect of limestone use on the porosity and related properties of cementitious mixtures (cement paste, mortar and concrete), the overall assessment of the published data suggest that essentially there are two opposing phenomena of : (i) particle packing decreasing porosity and (ii) dilution effect increasing porosity at play with the former dominating at the initial introduction of limestone up to an optimum level within a range of 12-20% and the later effect taking over thereafter.

4. ANALYSIS OF PUBLISHED DATA

Table 3 shows that widely different test methods and procedures have been employed in studying the effect of Portland limestone cement on the pore structure of concrete and that this is considerably more so in the measurement of porosity than absorption and sorpitivity. The Porosity test methods have been separated for clarity into four main groups:

(i) Environmental scanning electron microscope, backscattered electron imaging, x-ray diffraction and nuclear magnetic resonance.
(ii) Model of Powers and Brownyard, hydration kinetics model and computer based model HYMOSTRUC.
(iii) Mercury intrusion porosimetry (MIP).
(iv) Liquid displacement (LD).

This examination also suggested that MIP and LD are the two most reported test methods and this is considered to be due mainly to the relative ease of use of these two methods and in the case of MIP the extent of experimental data produced. In regard the water absorption measurements, the absorption by immersion was shown to be the common used method, because of its availability and simplicity for use.

Although not a preferred choice because of the interferences arising from the presence of aggregates, including the formation of interfaces, most probably due to the ease of handling of the test specimens, the vast majority of the studies (76%) have relied on the use of mortar and concrete instead of cement paste as the test material. The selection of test specimens in the form of cubes, cylinders and prisms appeared to be influenced by the relevant Standard specifications **(Table 3)**.

With the exception of few, and to certain extent influenced by the local standard specifications, moist curing with relative humidity ≥ 90%, temperature 20-25°C have generally been adopted, although interestingly some studies have used lime saturated water for curing to prevent possible leaching and carbonation of the test specimens and few adopted R.H. at 65% and temperatures at 5, 37 and 40°C for curing of the test specimens. However, the curing duration was found to be varied greatly, where the period of 28-90 days used in more than half of the studies undertaken, followed by in decreasing order, periods of 7-21 days, 1-3 days and 120-360 days. Similarly, time after which measurements are taken for absorption and sorptivity tests was found to be varied, with up to 24 hr period most frequently adopted **(Table 3)**.

The most of porosity measurements have been provided in the form of capillary porosity and total porosity with one reporting the results in the form of gel porosity. The sorptivity results have largely been reported in the form of initial sorptivity that takes up to 72 hours to complete, with few extending the test to measure final sorptivity at the end of 7 day period. Although the majority of the preparation of the test specimens have been according to standard procedures, a significant minority of 27% do not cite such information.

Given the large number of variables involved in the form test materials, test methods and procedures used, as can be seen from **Table 3**, the effect of limestone on the porosity and porosity related properties of cementitious mixtures (in the form of cement paste, mortar and concrete) the reported data can be best analysed and evaluated in comparison to the corresponding Portland cement (used as reference). However, some of the relative values were considered numerically distant from the rest and therefore have been regarded as outliers and not considered further in the analysis.

4.1 Porosity

As the cement paste in concrete is the only chemically active component, and it also contains the majority of the pores **[47]**, it is reasonable first to study the influence of limestone on the porosity of cement paste before dealing with the results obtained with mortar and concrete mixes.

The published results obtained using the two most popular methods, MIP and LD, are shown plotted in **Figure 1 (a and b)**, respectively.

It is clear that data population is limited and efforts to model the results using straightforward linear, logarithmic and polynomial trend lines proved to be unsatisfactory, yielding correlation coefficient (R^2) values for example below 0.40 (solid lines).

Notwithstanding the above, the observation of the data tend to suggest that for practical purposes limestone addition of up to 20-30% may not significantly affect the porosity of the cement paste, but thereafter the porosity may increase linearly with further increase in the limestone content (broken line). The data for cement paste reported in the literature for all the test methods as shown plotted in **Figure 1 (c)** tended to confirm the above, possibly with potential for some improvement in the porosity of cement paste with the use of limestone up to 30%.

The data plotted from the mortar/concrete studies in **Figure 2** suggest the limit of up to 20%. Thus, on the basis of **Figure 2**, it would appear that:

(i) Limit on the addition of limestone in cementitious mixtures in the form of cement paste **(Figure 1)** reduces when aggregates are introduced to form mortar/concrete mixtures **(Figure 2)**. In this case, as a way of example, a limit of 30% addition of limestone with cement paste **(Figure 1c)** was found to reduce to 20% limestone addition with mortar/concrete **(Figure 2)**.

(ii) The limestone addition as component of cement up to 35% as per specification of CEM II/B in **BS EN 197-1:2011 [22]** for common cements for use in concrete may perhaps be too high and that a figure of about 20% (CEM II/A) could be more appropriate and/or safer, depending upon the design strength and durability requirement of concrete.

4.2 Water Absorption and Sorpitivity

Little research has been reported on the effect of limestone addition on water absorption and sorptivity of cement paste and the data pertaining to these aspects couldn't be analysed. Thus, the data published for mortar and concrete are analysed as shown plotted in **Figures 3 and 4**. The scatter in the reported data is understandable given that the procedural differences particularly the duration of curing and measuring time have been varied greatly in the widely sourced data.

Figure 3 (a and b) for absorption and sorpitivity respectively show that in essence the two properties are affected similarly showing that limestone addition may be used without adversely affecting the two properties of the concrete. However, the two methods, in a limited manner, give different limiting values for limestone addition; 17.5% for absorption with coefficient of correlation (R^2) at 0.755 **(Figure 3a)** and 25% for sorpitivity with much reduced R^2 at 0.496 **(Figure 3b)**. Analysing the two sets of data together with the porosity results **(Figure 2)** produced a limiting value of 15% for limestone addition, **Figure 4**.

4.3 Compressive Strength and Porosity Relationship

Compressive strength, considered as the most important property of concrete, with cement pastes has been shown to relate directly to the volume and distribution of pores (porosity), **[125]**. In order to study the relationship between the strength and porosity of PLC cement paste and mortar/concrete mixtures, the published data taken from the publications that were sourced to study the effect of PLC addition on porosity, absorption or sorpitivity, have been analysed to evaluate the strength results relative to limestone addition, as shown plotted in **Figure 5**. This shows limits on PLC content of 25% and 17.5% for cement paste and mortar/concrete mixtures respectively, beyond which strength could be expected to decline with increasing PLC addition. These trends are similar to those observed for porosity of cement paste and mortar/concrete mixtures **(Figures 1c and 2)**.

Additionally, and for comparison purposes, the strength data from other publications, but limited to a period of ten years 2005-2014, that have not been included in the main study have been analysed separately to compare and confirm the effect of PLC on strength. This data is shown plotted in **Figure 6** and generally shows a similar trend for the effect of PLC addition on strength, albeit with slightly lower limits on PLC content and the difference between the cement paste and mortar/concrete limits less marked.

The effect of limestone on (i) porosity, and porosity related properties such as absorption and sorpitivity, and (ii) compressive strength are shown plotted for (a) cement paste mixtures and (b) mortar/concrete mixture in **Figure 7 (a) and (b)** respectively. This Figure is based on the published data in the literature sourced globally and in this respect whilst the data population shows high variability it helps to underpin the reported study. The two figures are based on total data populations compiled for the cement paste and mortar/concrete, and the two messages emerging in a way of confirmation, are:

- Limestone addition can be adopted for use as a component of cement up to a limited level beyond which the quality of cementitious mixtures in the form of cement paste and mortar/concrete in the hardened state will decline.

- The introduction of aggregates in a cementitious mixture reduces the limit on limestone content that can be applied without adversely affecting their quality.

In addition, based on **Figure 7** the correlation between the porosity related properties and strength development of PLC can be presented as in **Figure 8**. This shows the inverse relation between the two, where the increment of the pore volume leads directly to decline in the compressive strength. This agree with what have been reported in a previous study **[99]**.

4.4 Factors Affecting the Pore Structure of PLC Mixtures

This section considers other parameters that may influence the limestone addition effect on the pore structure of concrete, as follows:

4.4.1 Type of Portland Cement and Limestone

A limited number of studies have reported on the effect of the chemical composition of Portland cement and limestone on the porosity and sorpitivity of cement paste **[33]** and concrete **[102, 106, 135, 157]**. Given the simplified expression used for the complex Portland cement chemistry, for example in the form of C_3S, C_2S, C_3A and C_4AF, which can easily lead to large variations with small changes in the commonly measured oxide composition and likewise the inability to define finely the compositional make up of limestone, the PLCs can vary easily with minor changes in the composition of the PC and limestone. Such difficulties in using the data, arising from different sources, make it difficult to establish a definitive correlation between the state of the pore structure of PLC and its chemical composition. Not withstand this, the main findings of each of the reported studies are summarised as follows:

- A study of porosity and sorptivity of cement paste of two Portland cements having different C_3A content used with limestone addition showed no clear trend for the effect on the performance of PLC and no conclusion could be drawn between the amount of C_3A in the PLC and its pore structure **[33]**.

- Two types of Portland cement and limestone each, have been tested and no considerable difference was observed between the microstructure of the resultant cement pastes. Even so, the cement with lower porosity was attributed to its higher C_3S/C_2S ratio giving rise to higher rate of hydration, where more hydration products were formed at the studied ages (2 and 28 days) and denser microstructure obtained. Whereas the change in limestone type (chemical composition) was considered not significantly affect the porosity of the blended cement **[135]**.

- An investigation of sorptivity of PLC concrete with two different clinkers and three types of limestone concluded that depending on the PC composition and the cement fineness, there is an optimum limestone content at which the sorptivity of concrete is minimum. Although not stated, the analysis of the reported data showed the optimum limestone content to be about 15% in this case **[157]**.

- Two different samples of commercially available PLC (CEM II/A-L 42.5) were tested and no significant effect on the sorptivity of the produced concrete was reported **[102]**. However, the observation of the chemical composition of the two cements used in this study found to be essentially similar.

- The effect on porosity and sorpitivity of concrete of limestone addition using five different Portland cements and limestone samples at 5 and 25% addition levels were examined. Although differences in the measured results up to 10% with limestone type were reported, no explanation have been put forward, probably the differences were considered to be insignificant **[106]**.

4.4.2 Method of Producing and Fineness of PLC

Two studies **[58, 157]** involved two different methods of producing PLC, namely inter grinding and blending, with limestone content at 10% or 20%. The fineness of the PLC varied from 3640 to 5980 cm^2/g. The concrete mixes were prepared using a constant water/cement ratio of 0.5 or 0.65 and water cured at 20°C for 28, 90 and 150 days. The results showed that, for all intense and purpose, pore structure of concrete measured in the form of porosity and/or sorpitivity is:

(i) For a given fineness of PLC not significantly affected, whether it is produced by inter grinding or blending.
(ii) For a given concrete in terms of its water/cement ratio, curing and age at test, the fineness of PLC can be expected to lead to some improvement in its pore structure, through better particle packing achieved.

4.4.3 Curing

The data on the effect of curing on the pore structure of cement paste and concrete incorporating Portland limestone as component of cement is limited **[40, 106]**. Indeed, it is also cursory as one of the studies reported the use of only 5% limestone which is within the permissible limit for minor additions to Portland cement clinker in **BS EN 197-1:2011 [22]**. As to be expected, PC and PLC with the moist storage produced almost similar performance, with respect to curing temperature ranging from 5 to 40°C and duration of curing up to 180 days. The results of the other study which used cements with 0%, 5% and 25% limestone content in concrete having water/cement ratio of 0.6 and curing it in water and air for 28 days showed higher value for porosity and sorpitivity at 25% limestone content with water curing and much more higher with air curing.

4.4.4 Maturity and Water/Cement Ratio

Figures 9 and 10 have been developed based on the results obtained from the published literature that studied the influence of maturity and water/cement ratio on the effect of limestone addition on the pore structure of cementitious mixtures in the form of cement paste, mortar and concrete. In developing these Figures, the results of porosity, absorption and sorpitivity have been considered together to reflect pore structure of the cementitious mixtures and expressed in relative terms to the corresponding mixes without limestone. Furthermore, to eliminate the effect of curing only the data obtained with water curing, and moist curing with relative humidity greater than 90%, have been used.

Figure 9 shows that PLC concrete is noticeably more sensitive to moist curing than PC concrete and makes three useful points for developing the use of limestone as cement component in concrete:

- PLC concrete would require some initial moist curing in order to develop a pore structure of similar denseness to that of the corresponding PC concrete. Apart from strength, as sound pore structure of concrete is critical in developing its general resistance to deterioration, curing factor should be considered carefully when deciding on the use of limestone as a component of cement in specifying concrete.

- Although the duration of moist curing requirement for PLC concrete to match the pore structure of PC concrete increases with limestone content, it exceeds one month with limestone content in the region of 25%.

- The rate of improvement in pore structure of PLC concrete with moist curing increases with limestone content, but nevertheless it is unlikely that above certain limestone content in the region of 25% the PLC concrete will ever, within a reasonable time-scale, develop pores structure of denseness similar to that of PC concrete.

In developing **Figure 10**, because of the limited data, pore structure related measurements such as porosity, absorption and sorpitivity taken at 28 days with water curing or moist curing with relative humidity greater than 90% have been used. This Figure shows how the use of limestone as proportion of cement content may influence the changing pore structure with water/cement ratio and makes the following main points:

- Whilst, as to be expected, the pore structure of concrete improves with decreasing water/cement ratio, in relative to PC concrete this effect is also influenced by limestone addition.

- The rate of the above influence may generally be constant with low water/cement ratios up to about 0.40 and thereafter the pore structure is adversely affected at an increasing rate with increasing water/cement ratio and increasing limestone content.

- The maximum limestone content with moist curing of 28 days is about 25% at water/cement ratio of between 0.45 and 0.50. Higher water/cement ratios up to 0.70 may be used if limestone content is reduced.

4.5 Limestone with other Additions

As the new national and international Standards such as **BS EN 197-1:2011 [22]** and **BS EN 206:2013 [23]** are accepted in practice, the use of composite cements can be expected to grow. The information available on this aspect of limestone addition is summarised in **Table 4** from which the following main points can be ascertained:

- **Limestone in combination with fly ash**: It can improve the pore structure related properties such as porosity, absorption, and sorpitivity, of cement paste, mortar and concrete. However, this improvement is limited up to a certain level of addition beyond which the opposite happens.

- **Limestone in combination with GGBS**: This effects in the same manner as fly ash and depending upon the addition level it can be beneficial.

- **Limestone in combination with Silica fume**: When properly used, its use improves the pore structure related properties of the end product (in the form of cement paste, mortar and concrete).

- **Limestone in combination with Metakaoline**: Its effect is similar to silica fume.

Table 4 also suggests that the beneficial effect of these additional materials is generally realised through the improvement in the processes of hydration, linked with the fineness and chemical composition, in particular the alumina content leading to the additional carboaluminate hydrates, and thereby creating a denser cement paste **[124]**.

The variable effect has been observed with the use of fly ash and GGBS, where the enhancement of the pore structure was present until certain limit and thereafter the volume of the pores showed increase due to the dilution of Portland cement.

Although few studies have reported that the use of additions to Portland limestone cement to form composite cements adversely affect the pore structure of the resulting product, this has been observed to be due to improper use of the additions such, as using silica fume without a water-reducing admixture **[5]**.

5. IMPROVING PLC PERFORMANCE IN PRACTICE

Although the Standards allow up to 35% limestone addition in the cement for making concrete, global data analysed in this study suggest that PLC with limestone content greater than 15 to 25% may adversely affect the porosity of concrete, and thereby its overall performance in structures and in terms of strength the maximum amount of limestone that can be considered a safe use is more likely to be about 15%. However, this situation can be improved by reducing the water demand of a concrete mix as suggested in a previous study **[45]**:

(i) Optimising particle packing by revising the proportions of course and fine aggregates, and/or introducing the use of fillers.

(ii) Adopting the use of water-reducing admixtures, particularly the polycarboxylate ether (PCE) based products.

(iii) Developing the use of limestone with other additions such as small proportions of silica fume and metakaolin, as mentioned previously.

Such applications are being successfully adopted in the use of concrete in practice and in this case could help to develop a greater and more assured outlet for the use of limestone in concrete and at the same further improve its pore structure and thereby its general performance in terms of engineering properties and durability performance.

6. CONCLUSIONS

Based on the review of the literature and the analysis and evaluation of the data published since 1993, the following conclusions are drawn:

(i) The pore structure related properties (porosity, absorption and sorptivity) of cementitious mixtures (paste, mortar and concrete), remain unimpaired up to a maximum 25% addition of limestone to Portland cement, and beyond this threshold the pore structure of the PLC would begin to deteriorate, which for practical purposes can be assumed to take place at a constant rate with increasing limestone content. The limestone addition levels vary in national and international Standards, ranged from 10% to 35% **(Table 1)**.

(ii) Although the addition of limestone to Portland cement does not affect the relationship between the pore structure of the end product and its strength, the limit on limestone content for the strength to remain unchanged is likely to be less than that for the pore structure.

(iii) The variation in the chemical composition of Portland cement and limestone does not show a clear relationship with porosity, water absorption and sorpitivity of cement paste or mortar/concrete.

(iv) Combination of Portland cement and limestone after grinding separately or by inter grinding show no notable change on the pore structure properties, whereas a significant increase in the fineness of PLC can lead to some reduction in the porosity of the end product in the form of cement paste, mortar and concrete.

(v) The effect of curing temperature on pore structure of PLC paste at 5°C to 40°C, showed that, similar to Portland cement, its pore structure is adversely affected with increase in temperature. However, as only one study was found in the literature, this cannot be considered as a definitive conclusion, particularly when limestone content used in the study was similar to the permissible limit for minor additions in the Portland cement.

(vi) Similar to Portland cement, water curing of PLC concrete was found to improve its pore structure in comparison to air curing.

(vii) The condition of the pore structure of cement paste, mortar and concrete made with PLC improves with age (maturity), particularly during the first seven days, after which the improvement progresses steadily up to the age of 28 days and beyond.

(viii) To achieve comparable pore structure to Portland cement with 28 day moist curing at water/cement ratio of between 0.45 and 0.50 limestone content in PLC should be limited to a maximum value of 25%.

(ix) The composite mixture of Portland limestone cement with other additions may show a complementary effect on the pore structure of cement paste, mortar and concrete. However, this improvement is limited to a certain level of addition beyond which the opposite happens.

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List of Table Headings:

Table 1: Limestone contents permitted in Portland limestone cement in some national and international standards world-wide.

COUNTRY	LIMESTONE CONTENT, %	STANDARD/SOURCE
(a) Standards use 35% maximum limestone addition level.		
UK and Europe	CEM II/A: 6 to 20 CEM II/B: 21 to 35	BS EN 197-1:2011 [22]; EN 197-1:2011 [52]
South Africa	CEM II/A: 6 to 20 CEM II/B: 21 to 35	SANS 50197-1:2013 [140] (based on EN 197-1:2011)
Singapore	CEM II/A: 6 to 20 CEM II/B: 21 to 35	SS EN 197-1:2014 [137]
Mexico	6 to 35	NMX-C-414-2010 [105]
(b) Standards adopt maximum limestone addition level below 35%.		
USA	>5 to 15	ASTM C 595-M-2015 [8]
	>5 to 15	AASHTO M240–2015 [7]
Canada	>5 to 15	CSA A3001-2013 [27]
Australia	8 to 20	AS 3972-2010 [143]
New Zealand	up to 15	NZS 3125:1991 (Amended in 1996) [144]
China	up to 15	[69]
Iran	6 to 20	[122]
The former USSR	up to 10	[146]
Argentina	≤ 20	[146]
Brazil	6 to 10	[146]
Costa Rica	≤10	[146]
Peru	≤15	[146]

Table 2: Summary of the published literature findings concerning the effect of limestone addition[a,b].

TYPE OF EFFECT		MAJOR SUGGESTED CAUSES		
		Porosity	Absorption	Sorptivity
Higher	**Publications**	**24 %**	**32 %**	**33 %**
		LS particles do not expand in the matrix as PC does. Dilution effect on PC since the LS is inert filler. Higher w/c due to the reduction of PC content. LS particles have a greater critical pore diameter than PC. No clarification.	Increased permeable pore space. Formation of coarser pores. No clarification.	Higher porosity. Higher permeability. Porosity is reduced at the expense of capillarity and sorptivity. No clarification.
Lower	**Publications**	**39 %**	**22 %**	**25 %**
		Enhanced particle packing. Larger hydration products. Heterogeneous nucleation. Lower heat of hydration. Decrease in the water of consistency of the cement paste. No clarification.	Less number of capillary pores. Finer pores. Improvement in the distribution and tortuosity of the pores system. No clarification.	Less degree of interconnectedness of pores. Smaller capillarity pores. No clarification.
Variable	**Publications**	**15 %**	**30 %**	**38 %**
		Decreases with improved PSD until optimum level (15-18%) and then increases due to dilution of PC. Extent of fineness of particles. No clarification.	Decreases with improved PSD until optimum level (12-15%) and then increases due to dilution of PC. No clarification.	Decreases with improved PSD until optimum level (15-20%) and then increases due to dilution of PC. Fineness, type of grinding and type of blending with PC. No clarification.
No change	**Publications**	**2 %**	**0 %**	**4 %**
		No clarification.	-	No clarification.
Unclear	**Publications**	**20 %**	**16 %**	**0 %**
		No reference PC mix. No results and statements in regard.	No reference PC mix.	-

[a] Note: LS=Limestone, PC=Portland cement, PSD=Particle size distribution.

[b] **Data taken from** [4-6, 9-11, 13, 14, 17, 18, 24-26, 29, 30, 33, 36-43, 46, 48-51, 54, 56-63, 65, 68, 70-74, 76, 77, 80, 82, 84, 88, 89, 92-97, 99-104, 106-117, 119, 120, 122-124, 128, 129, 131, 133, 135, 152, 153, 156-161, 169-171].

Table 3: Details of test methods undertaken to study the effect of limestone addition on the pore structure[a].

PARAMETER	MEASURMENT		
	Porosity	Absorption	Sorptivity
Test method	Electron microscopy and microstructural imaging:{Number of publications =11} Numerical and computer models: {8} Mercury intrusion porosimetry: {25} Liquid displacement: {19}	Water absorption by immersion: {16} Capillary rise: {4} Initial surface absorption: {2}	Capillarity: {28}
Material	Hydrated cement paste: {31} Mortar/Concrete: {32}	Hydrated cement paste: {2} Mortar/Concrete: {20}	Hydrated cement paste: {1} Mortar/Concrete: {27}
Specimen	Cube: {36}, Cylinder: {7}, Prism and other: {20}	Cube: {13}, Cylinder: {3}, Prism: {6}	Cube: {13}, Cylinder: {13}, Prism: {2}
Curing	Ordinary water: {43}, Air curing {1} Lime saturated water: {6}, Moist: {12}	Ordinary water {13} Lime saturated water: {2}, Moist: {7}	Ordinary water: {21}, Air curing {1} Lime saturated water: {3}, Moist: {3}
Temperature	5°C {1}, 20°- 25°C {61}, 37°- 40°C {2}	20°- 25°C {22}	20°- 25°C {28}
Relative humidity	≥ 90% {61}, 65 % {1}	≥ 90% {22}	≥ 90% {27}, 65 % {1}
Curing length	1 - 3 days {13}, 7 - 21 days {18} 28 - 90 days {33}, 120 - 360 days {11}	1 - 3 days {5}, 7 - 21 days {6} 28 - 90 days {20}, 120 - 360 days {4}	1 - 3 days {2}, 7 - 21 days {6} 28 - 90 days {36}, 120 - 360 days {5}
Measuring time	--	10 minutes: {2}, 30 minutes: {4} 24 hours: {6}, 48 hours: {3} 72 hours: {1}, Not given: {6}	Up to 60 minutes: {6}, Up to 6 hours {2} Up to 24 hours: {14}, Up to 48 hours {3} Up to 72 hours: {3}, Up to 7 days: {2}
Type of outcome	Gel porosity: {1}, Capillary porosity: {30}, Total porosity: {32}	Water absorption: {22}	Initial sorptivity: {28}, Final sorptivity: {2}
Relevant Standard	Specified: {45}, Unspecified: {16}	Specified: {18}, Unspecified: {4}	Specified: {18}, Unspecified: (10)

[a] **Data taken from** [6, 9-11, 13, 14, 17, 18, 24, 25, 29, 30, 33, 36-43, 46, 48-51, 54, 56-61, 65, 71-74, 76, 84, 88, 89, 92-97, 99-104, 106-117, 119, 120, 122-124, 128, 129, 131, 133, 135, 153, 156, 157-159, 161, 169-171].

Table 4: Summary of the published literature findings concerning the effect of Portland limestone composite cement on the pore structure related properties[a].

TYPE OF EFFECT		MAJOR SUGGESTED CAUSES			
		Fly ash	GGBS	Silica fume	Metakaolin
Higher	**Publications**	**1**	**3**	**1**	**0**
		No clarification.	No clarification.	The increase in volume of pores could be attributed to the absence of a plasticizing agent in the studied ternary mixes, in which silica fume and limestone cannot act as effective fillers.	-
Lower	**Publications**	**3**	**6**	**3**	**2**
		Finer pore structure due the fineness of fly ash and limestone collectively. Hydration products resulting from fly ash reaction are able to fill the pores and participate in the formation of gel paste.	Fineness of the GGBS results in denser microstructure produced by lower calcium hydroxide content in which it has the effect of improving the hydration mechanism and the pore filling. Higher content of alumina in the mix (act as nucleation sites), limestone could participate in the hydration process by forming more carboaluminate hydrates, thus denser cement paste.	The pore packing and pozzolanic effects of silica fume results in a considerable reduction in the pore size and the connectivity of the cement matrix.	The addition of metakaolin increase alumina content in the whole mixture, where it allows more limestone to participate in the hydration reactions, creating additional carboaluminate hydrates.
Variable	**Publications**	**3**	**2**	**0**	**0**
		Decreases until certain level and then increases due to dilution of Portland cement clinker. No clarification.	Decreases until certain level and then increases due to dilution of Portland cement clinker. No clarification.	-	-
Unclear	**Publications**	**9**	**3**	**3**	**2**
		No reference PLC mix.	No reference PLC mix.	No reference PLC mix.	No reference PLC mix.

[a] **Data taken from** [1, 3, 5, 12-14, 20, 25, 30, 31, 36, 39, 41-44, 48, 55, 57, 60, 67, 72, 74, 75, 90, 91, 103, 113, 124, 132, 152, 161, 170, 171].

List of Figure Captions:

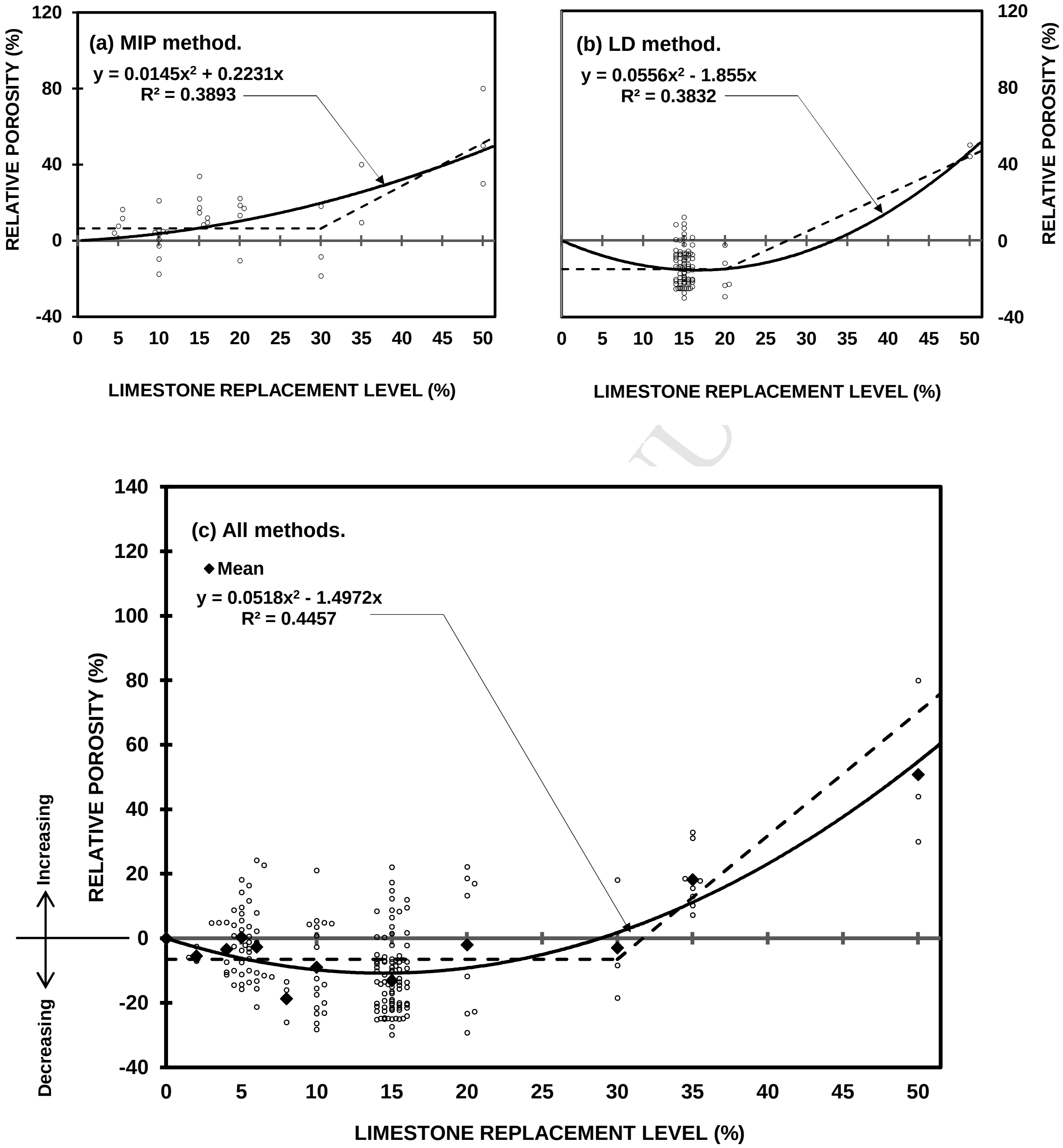


Figure 1: Limestone addition effect on porosity of cement paste mixtures using:

(a): MIP method, (b) LD method and (c) All methods.

Data taken from [9, 10, 17, 18, 24, 29, 33, 39, 40, 41, 43, 48-51, 71, 72, 84, 96, 97, 113,116, 117, 129, 135, 169, 171].

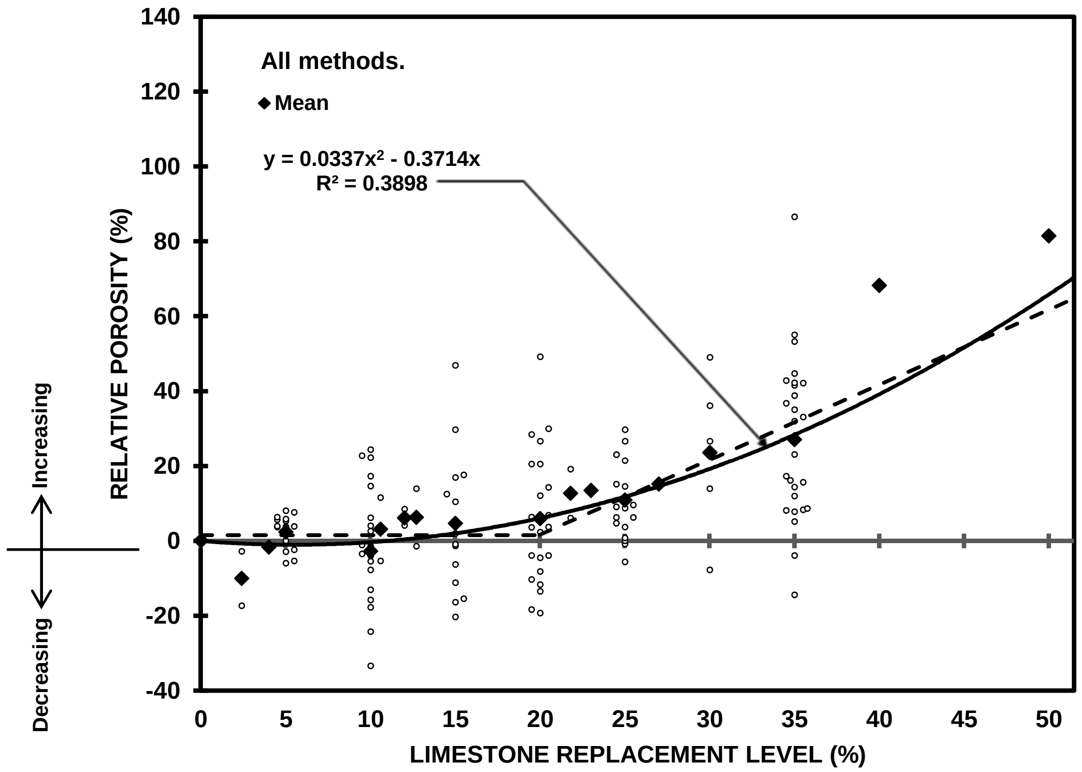


Figure 2: Limestone addition effect on porosity of mortar and concrete mixtures.

Data taken from [13, 14, 25, 36, 37, 42, 54, 56, 61, 71, 73, 76, 80, 88, 89, 93-95, 99, 100, 106, 108, 111, 112, 114, 115, 120, 124, 128, 131, 133, 156-160, 170].

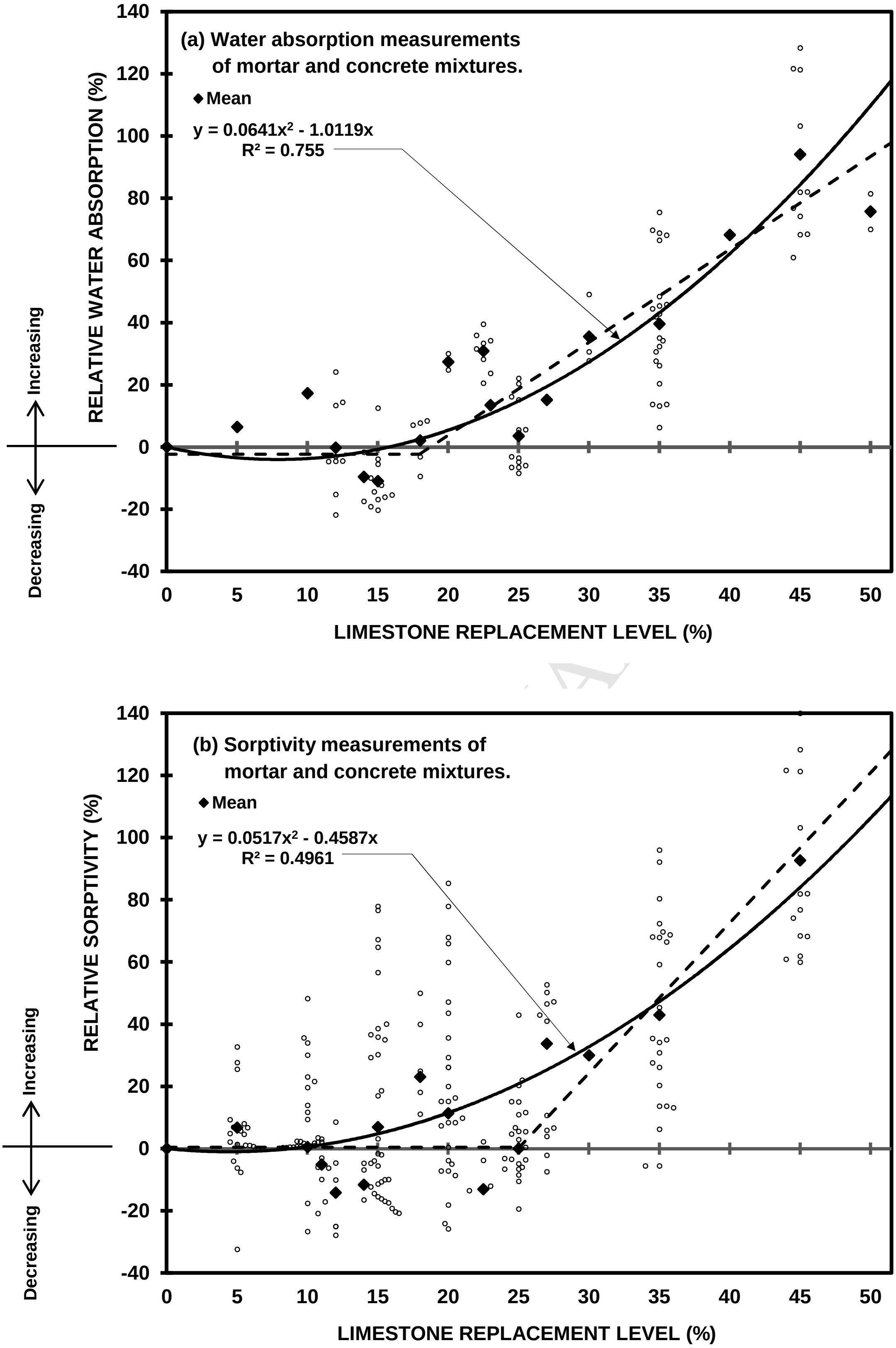


Figure 3: Limestone addition effect on

(a) Water absorption and (b) Sorptivity of mortar and concrete mixtures.

Data taken from [6, 25, 30, 33, 36, 37, 38, 46, 54, 57-60, 65, 74, 92, 99, 101-104, 106, 109, 110, 119, 120, 122, 123, 153, 156, 157, 159, 161].

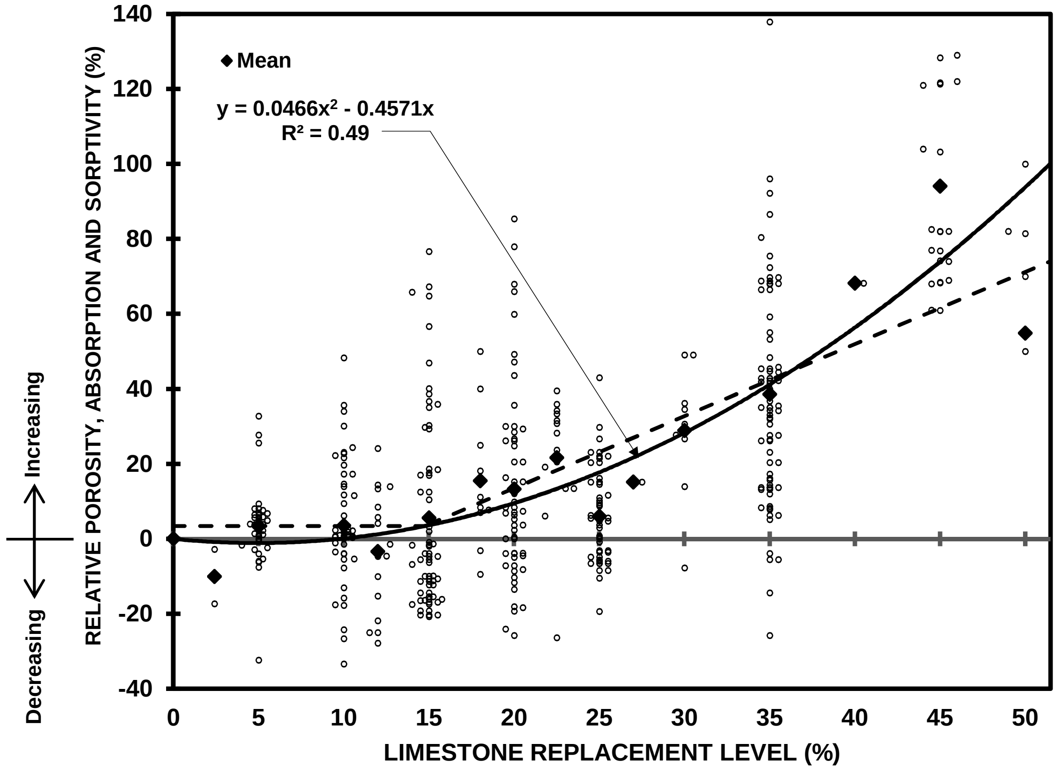


Figure 4: Limestone addition effect on porosity, water absorption and sorptivity of mortar and concrete mixtures.

Data taken from Figures 2 and 3.

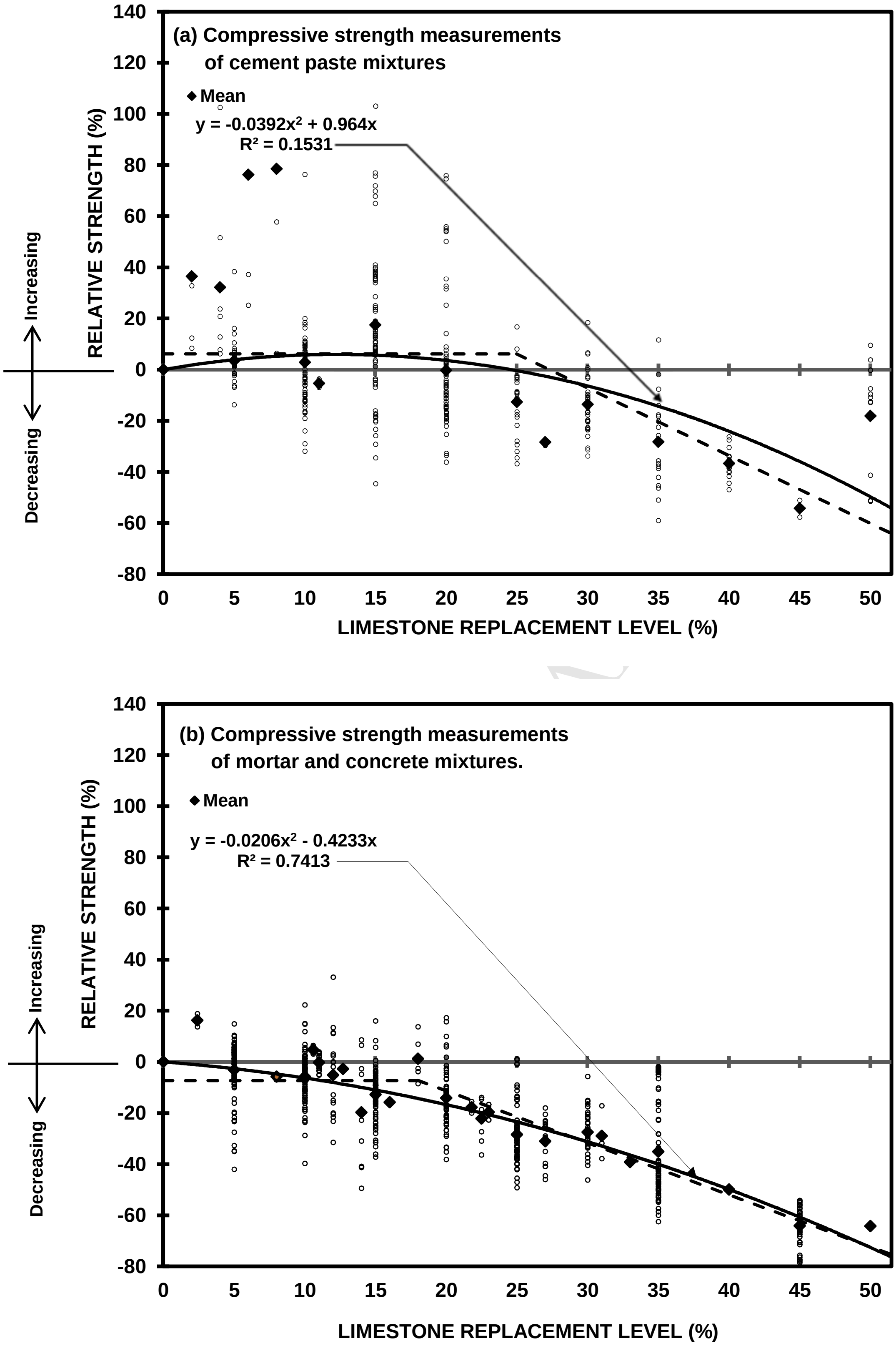
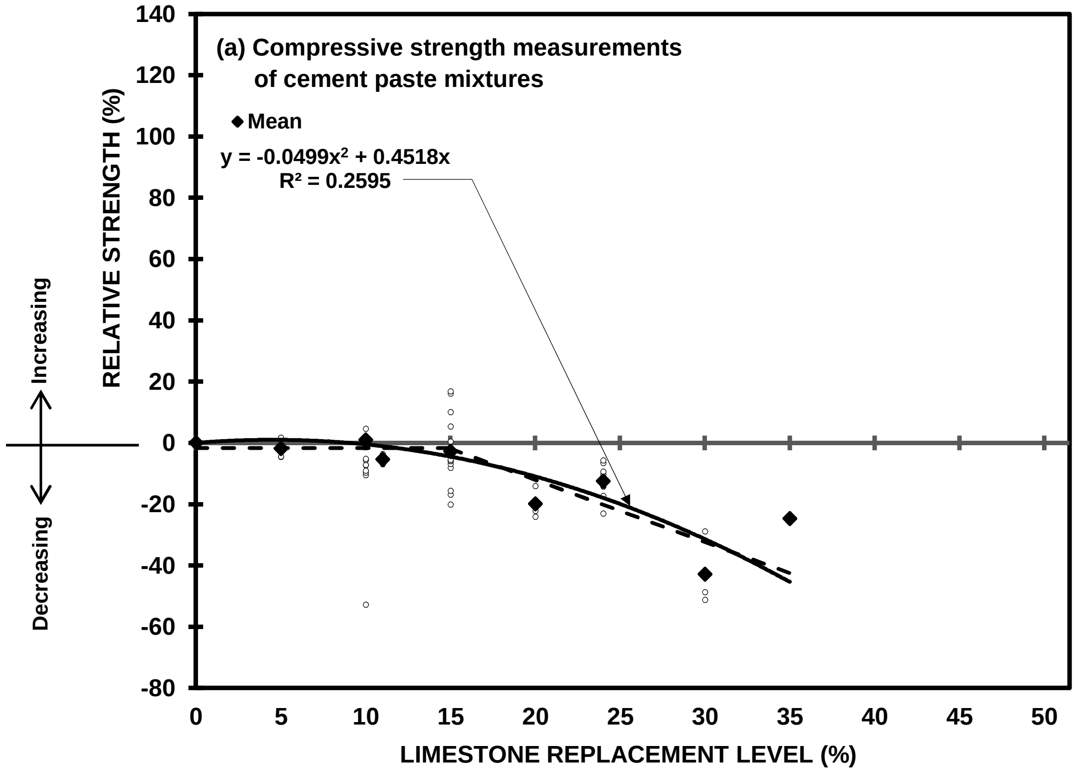


Figure 5: Limestone addition effect on compressive strength of (a) Cement paste and (b) Mortar and concrete mixtures.

Data taken from [4, 9-11, 13, 14, 17, 18, 24-26, 33, 36, 37, 39, 40-43, 48-50, 54, 56, 61, 62, 68, 71, 72, 73, 76, 80, 83, 84, 88, 89, 93-97, 99, 106, 108, 111, 112, 114, 115, 117, 120, 124, 128, 129, 131, 133, 156, 157-160, 170].



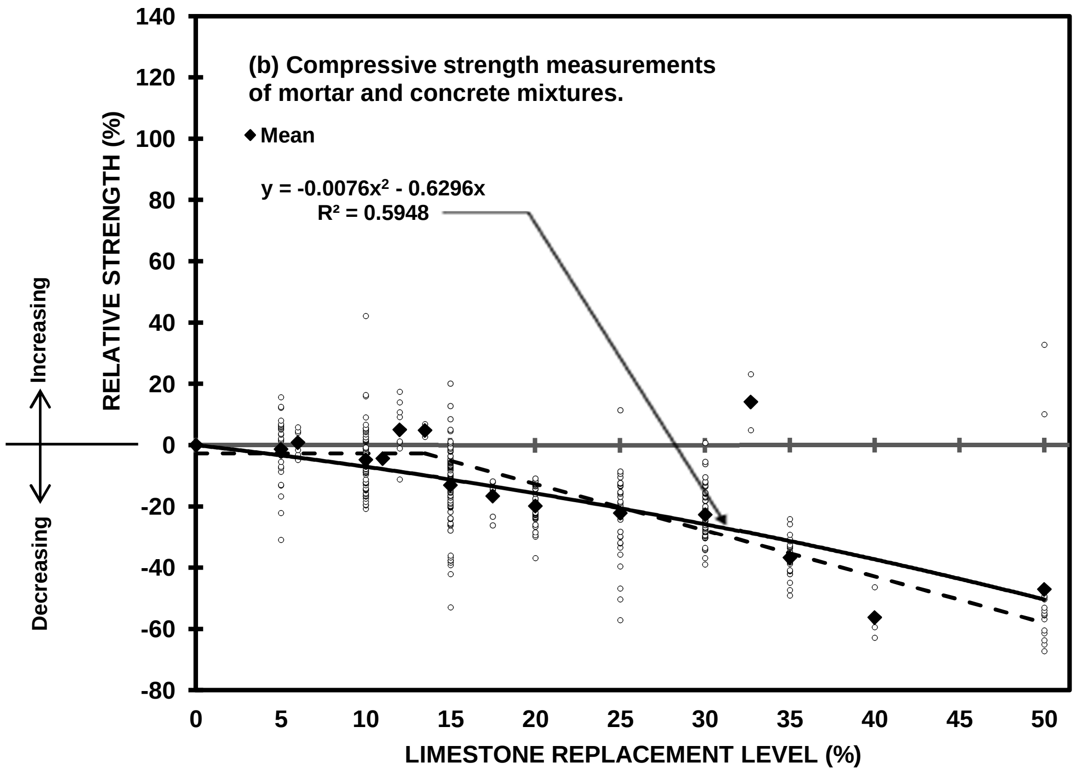


Figure 6: Limestone addition effect on compressive strength of (a) Cement paste and (b) Mortar and concrete mixtures for publications of years 2005-2014.

Data taken from [2, 16, 19-21, 28, 32, 34, 35, 44, 53, 64, 78, 81, 85-87, 90, 91, 98, 107, 118, 121, 126, 127, 134, 136, 137, 139-141, 145, 148-151, 154, 155, 162-168].

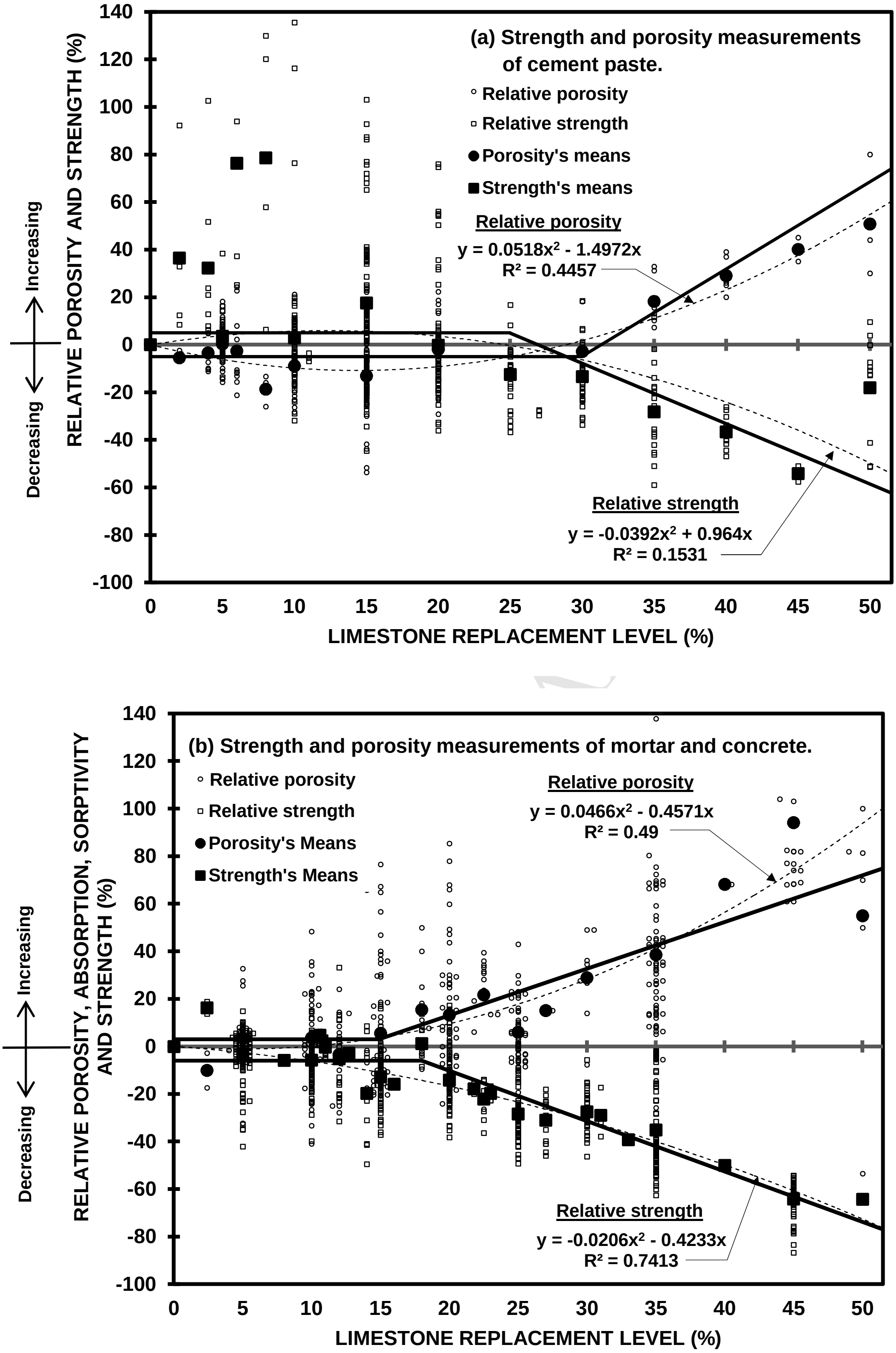


Figure 7: Effect of limestone addition as cement component on porosity and porosity related properties and strength development of (a) cement paste and (b) mortar/concrete mixtures.

Data taken from Figures 1, 2, 3, 4, and 5

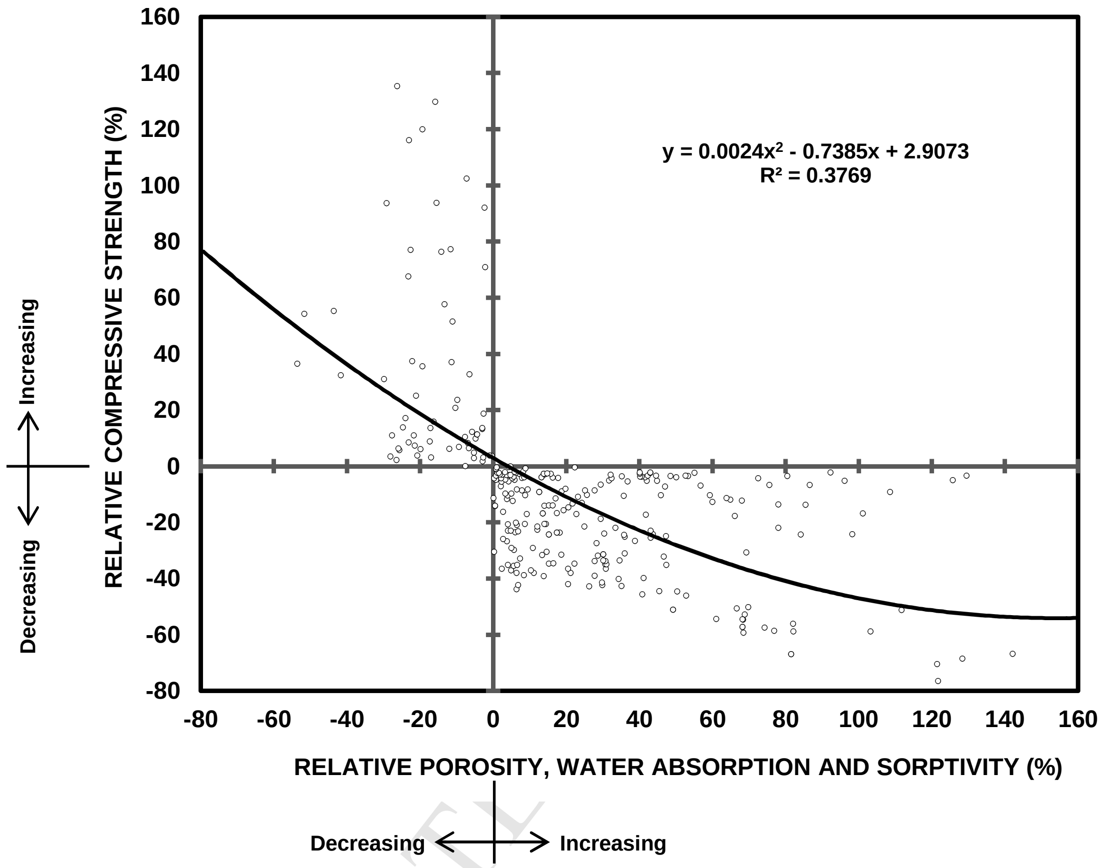


Figure 8: Strength verses porosity, absorption and sorptivity of cement paste, mortar and concrete mixtures.

Data taken from Figures 1, 2, 3, 4 and 5

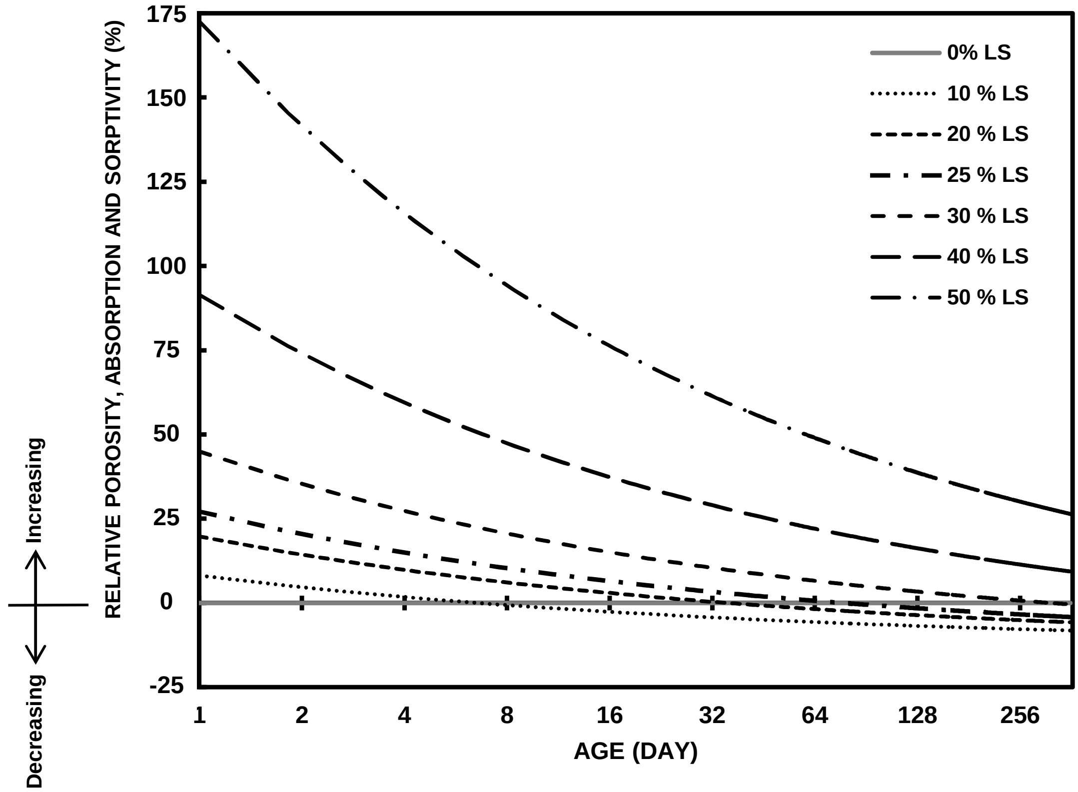


Figure 9: Relative porosity, absorption and sorptivity verses the age

This Figure has been developed based on the data in Figures 1, 2, 3 and 4

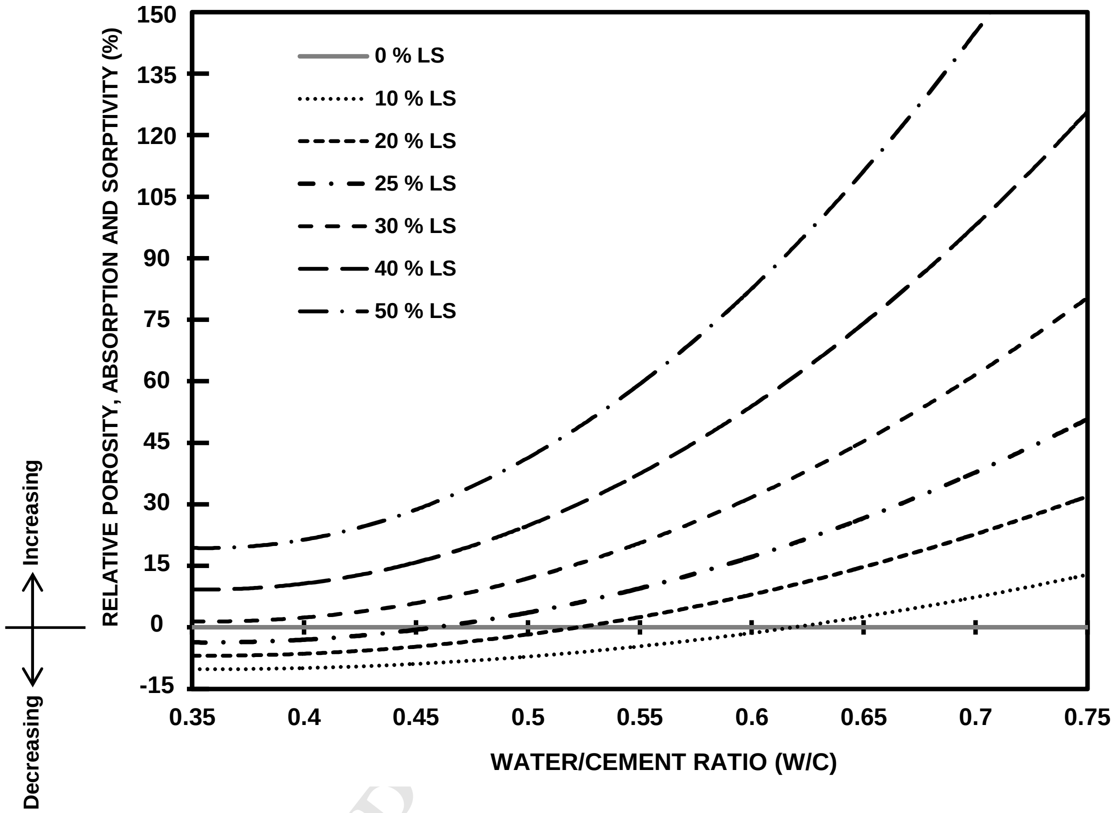


Figure 10: Relative porosity, absorption and sorptivity verses water cement ratio

This Figure has been developed based on the data in Figures 1, 2, 3 and 4

END