Towards a More Sustainable Australian Cement and Concrete Industry

James Mohammadi 1, Dr Warren South 2, Mr Des Chalmers 3
1, 2 Research and Technical Services, Cement Concrete and Aggregates Australia (CCAA)
3 Group Product Manager, Cement Australia

Abstract: This document provides a summary of results for the comprehensive test program to investigate the properties and performance of General Purpose (Type GP) cement prepared with up to 12% limestone content. An extensive literature review was carried out initially, and then an experimental research program designed. Cement, mortar, and concrete tests were conducted on a variety of mixes to evaluate the effect of introducing higher limestone mineral addition content to cement. The test results showed that there is no significant effect on most concrete properties due to increasing the limestone addition in cement. The result of this research supports the recommendation to increase the allowable limestone content of General Purpose (Type GP) cement to a maximum of 12%.

Keywords: General purpose (GP) cements; Limestone cement; Australian Standard AS 3972; Australian Standard AS 2350; Sulfate expansion; Rapid chloride penetration

1. Preface

The intent of the testing program was to study whether there is any detrimental effect on cement or concrete performance from the use of limestone mineral addition levels higher than the 7.5% allowed in AS 3972-2010 [1]. The upper limit chosen for this test program is 12%, based on the prevailing technical literature at the time the program was designed.

The program was designed and completed over 3 years (2011-2014) in two main stages under the supervision of Standard Australia BD-010 Working Group members. After the first stage of this research was completed, several members of the BD-010 Working Group sought to determine the effect of increasing limestone content in the cement used in a number of specific concrete systems. Therefore, an additional suite of tests was agreed among Committee members and carried out in 2014. The results of both research stages were documented and presented in two reports [2,3].

During the course of research program, combinations of different types of cementitious materials with plain Type GP cement were tested. Both plain cement and combinations of cement with supplementary cementitious materials (SCMs) were utilised to prepare cement paste, mortar, grout and different specific types of concrete samples. The research program included examining both laboratory and commercial mix designs. The suite of binder combinations included over two hundred different commercial concrete mixes, and involved standard laboratory mix designs, industrial grouts, normal and structural concrete grades, self-compacting, high-strength and durable concretes. The results of this research support the recommendation to increase the allowable limestone content of General Purpose (Type GP) cement to a maximum of 12%.

2. International Context for Cements with higher Limestone Contents

There is a strong international alignment for producing cement with higher limestone contents. For the past two decades European standards have permitted up to 20% and 35% limestone content for widely used CEM II types A/L and B/L, respectively. In a similar but later approach, Canada in 2008 and then in the USA in 2012 permitted higher limestone content in manufactured cement up to 15%.

The intent of this testing program is very clear. It is to determine whether there is any detrimental effect on cement or concrete performance from the use of limestone mineral addition levels higher than 7.5%, congruent with international practice.
3. Environmental Benefits of Cement manufactured with Higher Limestone Content

In the Australian context, the need to continue emissions reduction is clear. There is a long history of the use of supplementary cementitious materials in concrete in Australia. More recent data regarding the use of mineral additions in cement manufacturing are presented in Figure 1 (a). According to the Cement Industry Federation, gross emissions in 2010 were as shown in Figure 1 (b), at 660 kg per tonne of cementitious products, and further reductions have been seen in subsequent years [4]. A little over 25% reduction has been achieved since 1990. However, with community requirements influencing the use of alternative fuels, limited opportunity for thermal and electrical efficiency gains due to the relatively young age of cement manufacturing equipment, and little or no exploration of carbon capture & storage (CCS) strategies, clinker substitution remains the only viable alternative for emissions reduction. The most recent data, illustrated in Figure 1 (b), shows a continued downward trend in 2013/2014, coincident with the increase in allowable mineral addition in cement. To continue this trend, an increase in mineral addition in the most commonly used cements is required.

![Graph (a)](image1.png)  ![Graph (b)](image2.png)

**Figure 1:** (a) Use of mineral components in Australian cement manufacture 1990-2012, (b) CO\(_2\) emissions per tonne cementitious product produced, - Australia- 1990-2013 [4]

Overall, from an environmental point of view, intergrinding of limestone and clinker reduces clinker content of cement, saves energy and produces cement with lower embodied CO\(_2\) emissions. The use of mineral additions reduces the embodied emissions in cement roughly in proportion to the reduction in proportion of clinker in the cement. Increasing the mineral addition content of cement provides more sustainable Type GP cement, without compromising the existing structural and durability properties of concrete made with it.

4. Research Methodology

In practice, the achievable level of mineral addition and the material used as mineral addition will be determined by the cement manufacturer. Such decisions will be made with due reference to market requirements, cement manufacturing equipment capability and the availability of allowable materials. The cement Standard will prescribe the maximum level of allowable mineral addition in cements, but the actual level may vary from one cement manufacturer to another, as has always been the case.

More than 200 high-quality international published research documents were reviewed. Accordingly a research methodology was selected which is congruent with the American research “Specifications and Protocols for Acceptance Tests on Processing Additions in Cement Manufacturing” conducted by NCHRP [5]. The statistical evaluation involved three stages. Firstly, investigating any correlation between laboratory test results and limestone content of cement; secondly, checking the compliance of the test results with Australian standards and industry specifications; and, finally, investigating if any significant change in properties of cement, mortar or concrete is due to the increase in limestone content of General Purpose (Type GP) cement by conducting experimental inferential analysis.
5. Results and Discussion

5.1. Cement, Cement Paste and Mortar Results

In total, 43 binder combinations from different Australian manufacturers were prepared, using Type GP cement and flyash or slag. Limestones used were sourced from the mines used by each of the cement suppliers providing product for the trials. Selected limestones contained not less than 75% by mass of CaCO$_3$ as prescribed in the Australian Standard. Limestone with CaCO$_3$ content equal to or greater than 75% and less than 80% was acceptable provided the clay content determined by the methylene blue test, as given in EN 933-9, did not exceed 1.20%, and the total organic carbon (TOC) content, as given in EN 13639, did not exceed 0.50% by mass. For limestone with CaCO$_3$ content of 80% or greater, testing for clay content and TOC was not required. Cement samples with different limestone contents were prepared in plant trials carried out in the various cement plants providing product for the trials. This enabled an assessment of the performance of actual industrial product with increased limestone content.

Samples of cement were prepared with limestone mineral addition levels up to 12%. Thereafter, the effect of higher limestone on different properties of cement and cement paste, including chemical properties, setting time, hydration heat also compressive strength, drying shrinkage and sulfate expansion for mortar samples were examined.

All tests were performed in accordance with the relevant Australian Standard testing procedure in different industrial laboratories. The correlational analysis of data showed there is no significant correlation between properties of cement or mortar with limestone content of cement up to 12%. An insignificant increase (<3%) in 16-week sulfate expansion of some samples was observed which was statistically scattered and had $R^2$ of 3%. The only notable changes in characteristics were found for fineness. However, higher fineness and lower 45μ sieve residue were specifications set for the manufacturing process.

Results showed performance properties which were scattered within the usual statistical deviations arising from the number of test cements and binder combinations. Full cement and mortar correlational detail are available in the project reports. In addition to the conducted correlational study, test results for cement, mortar with higher limestone content were compared to the requirements of
Australian Standard AS 3972 as shown in Figure 3. All properties of 12% limestone content cement complied with the requirements of Australian Standards.

![Figure 3: Compliance with Australian Standards requirements (a) SO₄ content, (b) Chloride content, (c) Soundness, (d) Setting time, (e) Compressive strength, (f) Sulfate expansion and Drying shrinkage](image)

The third part of analysis included an inferential statistical analysis of cement and mortar test results. Considering the standard deviation, variation and distribution of test results, the statistical significance of test results was calculated. The majority of properties for cement and mortar samples prepared by higher limestone content did not show significant change from the samples prepared with control Type GP cement. The only statistically significant drawback found was a 5% reduction in mortar 28-day compressive strength, explained by the constraints of the test method, and congruent with reported international findings.

In summary, the results of cement, cement paste and mortar samples prepared with a higher limestone content demonstrated comparable properties with the control samples and there is no technical evidence that should limit the manufacture of Type GP cement with up to 12% limestone mineral content.

### 5.2. Concrete Results

The testing of the cements containing 7.5%, 10% and 12% limestone mineral addition was carried out in conjunction with relevant control cements as benchmarks. In addition to the 34 sets of standard laboratory concrete mixes using in-house mix designs, a wide range of concretes were prepared using current commercial mix designs for specific types of concrete. 52 sets of durable concrete; 24 sets of high-strength concrete; plus 35 and 32 sets of normal concrete grades N32 and N20 respectively were prepared. Moreover, an additional 33 sets of different specific types of concrete were provided from actual field concrete supplied for testing. Testing included those specified for fresh, hardened and durability properties namely (where applicable) concrete slump, concrete spread test, initial and final setting time, air content, bleed water, 1 day to 56 days compressive strengths, 56 day drying shrinkage, chloride ion penetration at 28 and 56 days, chloride ion migration, volume of permeable voids, creep test to 112 days, and modulus of elasticity at 28 and 56 days.
The evaluation of the data indicates there is little difference in performance between concrete using cement containing 12% limestone mineral addition and the control samples. This is supported by a statistical evaluation of the test results. Full correlational study, compliance with requirements, and inferential statistical evaluation details are available in the project reports. A summary of major findings is addressed as follows:

**Laboratory Standard Concrete**

The results of testing concrete based on standard laboratory mix design showed almost the same performance for 12% limestone mineral addition concrete samples and control samples. A statistically significant observed change was a reduction in bleed with increasing limestone content.

**Durable Concrete**

The test results for durable concretes indicated similar performance for 12% limestone mineral addition samples and control samples. The 12% limestone content durable concrete complied with all the benchmark requirements of this specific grade. Statistically significant changes were the reduction of bleeding and final setting time with increase in limestone content. Results for chloride ion penetration and chloride ion migration showed a significant improvement in concrete performance for the higher limestone content concretes.

**High-Strength Concrete**

Test results for high-strength concrete samples indicated similar performance for 12% limestone mineral addition samples and control samples. The 12% limestone content high-strength concrete complied with all the benchmark requirements of the high-strength specific grade. Similar workability and creep characteristics were observed for 12% limestone cement samples. Statistically significant observed changes were in the reduction of 1 day and 56 days strength by 7% and 1.6%, respectively. Test results for the modulus of elasticity at 56 days showed a 4% reduction compared to control samples.

**Normal Concrete Grades N20 and N32**

Test results for normal grade concrete samples showed almost the same properties for 12% limestone mineral addition samples and control samples. Both grades N20 and N32 prepared with 12% limestone content complied with all the benchmark requirements for these grades.

A statistically significant observed change for grade N20 was a reduction in bleed. No other significant change was detected.

A statistically significant observed change for grade N32 concrete was a reduction in bleed. In addition, relatively to the control samples, slump and air content results were 5% and 6% lower, respectively. However, higher strength values of about 3% to 6% for early ages (1, 3 and 7 days) test results were observed, which were statistically significant.

**Field Concrete**

Test results from field concrete samples exhibited similar performance for 12% limestone mineral addition concretes and control samples. The 12% limestone content concrete showed similar strength, drying shrinkage and fresh properties except for setting times, which were reduced by about 3-4%.

In general, it can be concluded that the test results for different specific types of field concrete did not show significant differences to control samples and supported the proposition of increasing the allowable mineral addition level in General Purpose (Type GP) cement, up to 12% limestone mineral addition.

**5.3. Supplementary Cement, Mortar, Grout and Concrete Results**

Several members of the BD-010 Working Group expressed concerned with the experimental program for the original test program (2011-2014). Specifically, they sought to determine the effect of increasing limestone content in the cement used in a number of specific concrete materials. Therefore,
Cement, mortar and concrete tests were conducted on a variety of mixes to evaluate the effect of introducing higher limestone mineral addition to cement. These were to be tested according to normal test methods used in Australia. The total number of prepared mixes was 61 which were distributed between different types of concrete and grout. There are major differences in the performance requirements and properties of these specific types of concrete. Therefore, it was decided to report the result of the studies for the different groups of specific types of concrete separately from the original program. In addition to the cement, cement paste and mortar testing, six specific types of concrete included grout, self-compacting, general purpose, durable, high strength and structural concretes were examined.

**Cement, Cement Paste and Mortar**

Some changes in chemical composition were noted due to a higher limestone content. In addition, there was a strong correlation between limestone content and surface area and the 45 μm residue due to the influence of the manufacturing specifications. There was no correlation between cement limestone mineral addition content and the compressive strength or drying shrinkage of mortar.

**Grout**

Grout test results include workability and strength results of three mixes prepared based on commercial mix designs. Results showed a significant improvement in the flow properties of grout made with cement containing the higher limestone contents in both the initial measurement and also after 45 minutes. The improvement may be due to the positive effect of higher limestone on the increase of water retention properties of grout. In addition, the better workability of mixes with higher limestone may be explained by softer and finer particles of limestone compared to the ground clinker particles. Literature supports the reported results for grout. Compressive strength tests were performed at different ages. Results revealed that there was no significant correlation between grout strength and limestone mineral addition content of cement.

**Self-Compacting Concrete (SCC)**

The higher limestone content of cement did not result in a significant change in concrete flow. The results from additional tests such as T₅₀₀ test and J-ring passing ability showed better workability of SCC with increased limestone content, however, the correlations were not strong.

![Figure 4: Self-compacting compliance with work-group requirements](image)

Considering results from all ages, slopes of trend lines and calculated values of R², it can be concluded that the increase in limestone content had no significant effect on the strength of SCC. SCC drying shrinkage results showed a slightly higher drying shrinkage for 12% limestone content, which may be due to the higher fineness of the cement, the increased limestone content or both factors. The correlations for chloride penetration and Volume of Permeable Voids (VPV) with limestone content of...
cement were assessed for the SCC mix series. No significant correlation between limestone mineral addition content and chloride penetration or VPV was found.

**Normal, Structural, Durable and High-strength Concrete**

In order to have a complete investigation regarding the effect of higher limestone on different types of concrete, 54 sets of concrete were prepared and tested by examining fresh, hardened and durability properties. It was found that the higher limestone content does not significantly affect the fresh properties of concrete. Results showed the air content decreased very slightly with the increase in limestone content. Moreover, the samples with increased limestone content did not have a significant effect on fresh concrete density or workability.

![Graphs showing changes in properties of concrete samples with different limestone contents](image)

**Figure 5: Relative change of properties of concrete samples prepared with different limestone contents (a) Compressive strength (b) Drying shrinkage (c) Chloride penetration ASTM 1202 method, (d) Chloride penetration NT 492 method (e) Volume of permeable voids AS1012.21 method**

The test results showed that at very early ages, the compressive strength of concrete increased slightly with the increase of limestone content. For other ages, the compressive strength of concrete samples was not changed by the increase of limestone content.

An insignificant increase in the drying shrinkage was observed for the majority of concrete samples at different testing ages (Figure 5). Comprehensive analysis of drying shrinkage test results revealed that slightly higher drying shrinkage was observed only for concrete samples prepared with a combination of Type GP cement and fly ash. The higher drying shrinkage for 12% limestone content is possibly due to the higher fineness of the cements containing the higher limestone mineral addition levels.

Durability test results revealed that there were negative correlations between chloride penetration and the Volume of Permeable Voids with limestone content of cement. These lower Volume of Permeable Voids and chloride penetration results suggest improved durability of concrete using cement with higher limestone content. The observed improvements were more significant for concretes prepared with Type GP cement, while there were mild or insignificant correlations found for those prepared by Type GP cement and fly ash.

It was found that for most properties no significant change was found for up to 12% limestone mineral addition content (Table 1). The only drawback observed with 12% limestone mineral addition was a
higher drying shrinkage measured at 21 days compared to the control samples. However, the measured concrete drying shrinkage at 56 days did not show any significant increase. In addition, it is important to take into account that the reported repeatability of the drying shrinkage test is 8% at the 95% probability level. The measured shrinkage for 12% limestone samples was 7% higher than the shrinkage of control samples.

Table 1: Overall evaluation of specific types of concrete test results for higher limestone cements

<table>
<thead>
<tr>
<th>Item</th>
<th>Correlation</th>
<th>Compliance</th>
<th>Inferential</th>
<th>Decision on addition</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Trend</td>
<td>Condition</td>
<td>10% content</td>
<td>12% content</td>
</tr>
<tr>
<td>Slump</td>
<td>neutral</td>
<td>insignificant</td>
<td>yes</td>
<td>yes</td>
</tr>
<tr>
<td>Air content</td>
<td>negative</td>
<td>insignificant</td>
<td>yes</td>
<td>yes</td>
</tr>
<tr>
<td>Mass per unit volume</td>
<td>neutral</td>
<td>insignificant</td>
<td>yes</td>
<td>yes</td>
</tr>
<tr>
<td>Strength 1day</td>
<td>positive</td>
<td>insignificant</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Strength 3day</td>
<td>neutral</td>
<td>insignificant</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Strength 7day</td>
<td>neutral</td>
<td>insignificant</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Strength 28day</td>
<td>neutral</td>
<td>insignificant</td>
<td>yes</td>
<td>yes</td>
</tr>
<tr>
<td>Strength 56day</td>
<td>neutral</td>
<td>insignificant</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Strength 91day</td>
<td>neutral</td>
<td>insignificant</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Shrinkage 21day</td>
<td>positive</td>
<td>insignificant</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Shrinkage 56day</td>
<td>positive</td>
<td>insignificant</td>
<td>yes</td>
<td>yes</td>
</tr>
<tr>
<td>Chloride ASTM 1202</td>
<td>negative</td>
<td>mild</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Chloride NT492</td>
<td>negative</td>
<td>mild</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>AVPV</td>
<td>negative</td>
<td>mild</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

The test results showed an improvement in the chloride penetration (ASTM 1202 Method) and Volume of Permeable Voids (AVPV) for concrete prepared with 12% limestone mineral addition. The improvement was significant and confirmed at a 95% confidence level. The better durability performance of concrete prepared with 12% limestone GP cement suggests the potential to enhance concrete performance or service-life and reduce maintenance costs.

6. Conclusion and Recommendation

The results of this project supported the proposition of increasing the allowable mineral addition level in General Purpose (Type GP) cement. It was found that by increasing limestone content of cement to 12% most properties of cement, mortar and concrete were effectively unchanged. There were generally very limited performance reductions and these were statistically insignificant. It is concluded that variability in performance among the different tests was influenced more by concrete mix design and binder composition than by limestone content in the Type GP cement.

The result of this research supports the recommendation to increase the allowable limestone content of General Purpose (Type GP) cement up to a maximum of 12%.

7. References

[1] AS3972-10, General purpose and blended cements, Sydney, 2010