How Does Pozzolanic Reaction Make Concrete "Green"?

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INTRODUCTION

When Portland cement is produced there is a significant amount of CO_2 emitted from the calcination of limestone. If the amount of CO_2 emitted can be reduced a system is considered to be "more green". Less cement in concrete would make concrete "more green". Leadership in Energy & Environmental Design (LEED) innovative credit ID-C1 provides a special credit for concrete if the amount of cement in a concrete mixture is reduced by 40 %(making the concrete environmentally green). One method to reduce the amount of cement in concrete is to use a pozzolan. How much cement can be reduced by using a pozzolan? Can a pozzolan alone produce a concrete with 40% less cement? If not, what other concrete mixture proportions should be controlled to achieve a 40% reduction (for example, reductions in water demand)? If a 40% reduction in cement is achieved is the concrete mixture at the optimum pozzolan percentage? What is the optimum pozzolan percentage? Does the optimum percentage vary with the pozzolanic index (reactivity of the pozzolan)? If a concrete mixture is not proportioned at the optimum pozzolan percentage is it truly green even if it provides a 40% reduction in cement? Could concrete be "more green" with less than a 40% reduction in cement content? The discussion in this paper endeavors to answer these questions.

POZZOLANIC REACTION—FLY ASH

If a concrete contains a pozzolan, less cement is required to obtain a specified strength. The amount of cement reduction will vary depending upon the reactivity of the pozzolan. A highly reactive pozzolan has more cementitious strength value than a lower reactive pozzolan. The amount of cement reduction will be greater with a more reactive pozzolan. What is pozzolanic reaction and how can it be measured? By definition a pozzolan by [1]ACI is: "a siliceous or siliceous and aluminous material that in itself possesses little or no cementitious value but that will, in finely divided form and in the presence of moisture, chemically react with calcium hydroxide (lime) at ordinary temperatures to form compounds having cementitious properties.

By definition to form a compound that has cementitious value a pozzolan must react with lime which in concrete comes from the cement. Two factors or assumptions must be defined. (1) Lime-- How much lime is available? (2) Pozzolanic Reaction--What is the chemical reaction of a pozzolan with lime?

1. Lime--$Ca(OH)_2$ Available

The amount of lime available is the lime produced during cement hydration and this will be reduced relative to the percentage of pozzolan, as discussed below. The amount of lime that is available will vary with cements. It will depend upon the amount of C_3S and C_2S in the cement. Each of these compounds react with water to form C-S-H(Calcium Silicate Hydrate) and lime-$Ca(OH)_2$. A small portion of this lime enters into reactions with cement alumina and sulfates to form compounds such as ettringite. Therefore not all of the lime-produced is available or free to react with a pozzolan. Various estimates of free lime can be found in the literature. These include 15-25% [2](Kosmatka, 2002), 20-30% [3](Popovics, 1998), approximately 22% [4](Massazza, 1988) ,etc. A value of 25% is assumed in the following discussion. That is 75% of the reaction products of cement by mass produces strength and the remaining 25% is free lime.

The amount of lime is also related to the amount of cement hydration. At early ages only a small amount of cement has hydrated and thus the amount of lime is low. For example if only 50% of the cement has reacted there will only be 50% of the potential lime available, for example 50% of 25% or 12.5% by mass of the reaction products of cement.

In addition, when a pozzolan is used, the percentage of pozzolan will reduce the available amount of lime. For example, a system that is 100% cement at full hydration will have 25% lime, and a system with 80% fully hydrated cement and 20% pozzolan will have only 20% lime available(80% *25=20%).

2. Pozzolanic Reaction

It will be assumed that all pozzolanic reactions are lime reactions with siliceous pozzolanic materials. Reactions with aluminous pozzolanic materials will be briefly considered below. The dominate hydration product of Portland cement is C-S-H (Calcium Silicate Hydrate). If it is assumed that the reaction of Pozzolan and lime produces the same hydration products as cement consider the following reaction:

$$\underset{222}{3\,[\,Ca(OH)_2\,]} + \underset{120}{2\,[\,SiO_2\,]} = \underset{342}{[\,3(CaO)\,2(SiO_2)\,3(H_2O)\,]} \qquad (1)$$

In this reaction 222 parts lime reacts with 120 parts silica for a mass ratio of 222/120 = 1.85. The value 1.85 is significantly higher than reported values. Values from 0.20 to 1.0 are reported by [4](Massazza, 1988). However, the reported values are based on the total mass of the pozzolan, which includes the reactive portion and the inert portion. If a pozzolan is only 25% reactive (75% inert, non-pozzolanic) and it combines with 0.46(%) of lime the reactive portion ratio is 0.46/0.25 = 1.84 (which agrees with equation 1). The ratio shown in equation 1 is assumed for the reactive portion and assumed for the development of the pozzolanic index that will be demonstrated.

In contrast to siliceous reactions, pozzolanic reactions could be between lime and aluminious pozzolanic materials. In this case the final hydration product of aluminous compounds in cement is assumed to be $3(CaO)Al_2O_3\,6(H_2O)$. Consider the following pozzolanic reaction

$$3\,[\,Ca(OH)_2\,] + Al_2O_3 + 3\,[\,H_2O\,] = 3(CaO)Al_2O_3\,6(H_2O) \qquad (2)$$

$$222 \quad + \quad 102 \quad + \quad 54 \quad = \quad 378$$

The relationship between lime and alumina is 222/102 = 2.18. Alumina combines with more lime than does silica.

The pozzolanic index used in this discussion is the decimal percent of the pozzolan (fly ash) that reacts at a defined age, for example 28-days. The index varies from 0 to 1. For example, an index of 0.25 indicates that 25% of the pozzolan has reacted.

CONCRETE MIXTURES-OPTIMIMUM POZZOLAN PERCENTAGE

The mass of reaction product produced by the reaction of pozzolan with lime can be calculated based on equation #1 above. If there is available lime in the system the amount of pozzolan (mass) that will react is $P_x * P_w$(P_w = mass-weight of pozzolan). The amount of lime that reacts is 1.85 $P_x * P_w$ based on equation 1. The total mass of pozzolanic reaction is the total of these two ($P_x\, P_w$) + (1.85 $P_x\, P_w$) = 2.85 $P_x\, P_w$. The mass of pozzolanic reaction is shown in equation 3.

$$\text{Mass of Pozzolanic Reaction} = (2.85\, P_x\, P_w) \qquad (3)$$

In the following discussion, $P_\%$ is the decimal percent of total cementitious material that is pozzolan. For example $P_\%$ for 400 lbs cement and 100 lbs fly ash is 100/(400+100) = 0.20 (20%)..

If pozzolan replacement of cement is 20% ($P_\%$ = 0.20), there is 0.80 parts cement and 0.20 parts pozzolan. The amount of lime produced from cement hydration is then (1-0.20)*0.25= 0.20 or 20% at full hydration. One(1) part silica reacts with 1.85 parts lime, therefore the lime available could react with 0.20/1.85 = 0.108 parts of silica. If the pozzolan index is 25% (P_x = 0..25) and the pozzolan percentage $P_\%$ = .20 (20%) the following is calculated. The reactive $P_x * P_\%$ = 0.25 * 0.20 = 0.05 (%) pozzolan (silica) is available to react which reacts with 1.85 * 0.05 = 0.0925 (%) lime of the 0.20 available. For an index of 0.25 the amount of excess lime (non-reacting) is 0.20-0.0925 = 0.1075 (%).

What is the optimum percentage for this pozzolan so that the reactive part of the pozzolan potentially reacts with all of the lime available? Consider the following equation:

$$(1 - P_{\%})*0.25/1.85 = P_X*P_{\%} \quad (4)$$

The left side of Equation 4 calculates the amount of silica that can react with the lime available at $P_{\%}$. The right side of the equation gives the amount of silica that is available, that is the amount of the pozzolan that is reactive, assumed to be reactive silica. Solving for $P_{\%}$ produces the following:

$$P_{\%\text{-OPTIMUM}} = 0.1351 / (P_X + 0.1351) \quad (5)$$

Using equation 5 for a pozzolan with an index of 0.25 (P_x = 0.25 at 28-days" age) calculates an optimum fly ash percentage of 0.1351/(0.25+0.1351) = 35.1%. If the pozzolanic index is a lower number the optimum fly ash percentages is higher. For a P_X of 0.15 the optimum percentage is 0.1351/(0.15 + 0.1351) = 47.3%.

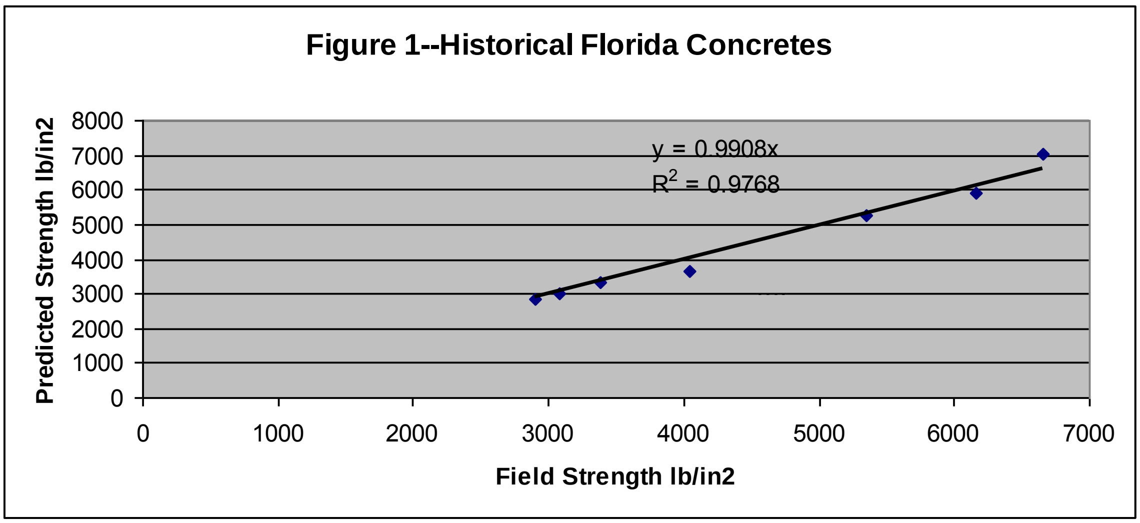
CONCRETE STRENGTH MODEL

The mix designs and compressive strength results demonstrated in this paper are based on a prediction model developed by this [5]author. The model is an updated version of the one previously published by the author. The model uses features of the [6]Powers gel space ratio and modifications for pozzolanic reaction. The model can very accurately predict concrete strengths as demonstrated in the following discussion.

The concrete mixes shown in Table 1 are historical Florida concrete mixes in that the same source of materials, cement, fly ash, sand, and coarse aggregate have been used in combination for more than 25 years. The strengths shown for these mixes are for field performance strengths. The strengths are averages of a minimum of 50 sets of concrete cylinders up to 200 sets of cylinders for each concrete mix. The majority of the tests were conducted late in 2004 and early 2005. The strengths shown are calculated average strengths for cylinders cast and tested by multiple testing labs, from multiple ready-mix plants and from multiple projects. This historical concrete field data is the same as reported in the 2006 paper by this author. The field concretes are compared to predicted model strength in Table 1. The model predicts concrete strength for these concretes with a correlation coefficient R^2 = 0.9768 as shown in Figure 1.

Table 1—Historical Florida Concretes versus Predicted Model Strength

Water lbs/yd^3	Cement lbs/yd^3	Fly Ash lbs/yd^3	Field Strength Lb/in^2	Predicted Strength Lb/in^2
270	285	140	2900	2830
270	300	140	3075	3010
270	325	140	3380	3320
270	350	140	4050	3655
270	450	140	5345	5255
270	500	100	6160	5930
270	550	100	6660	7060



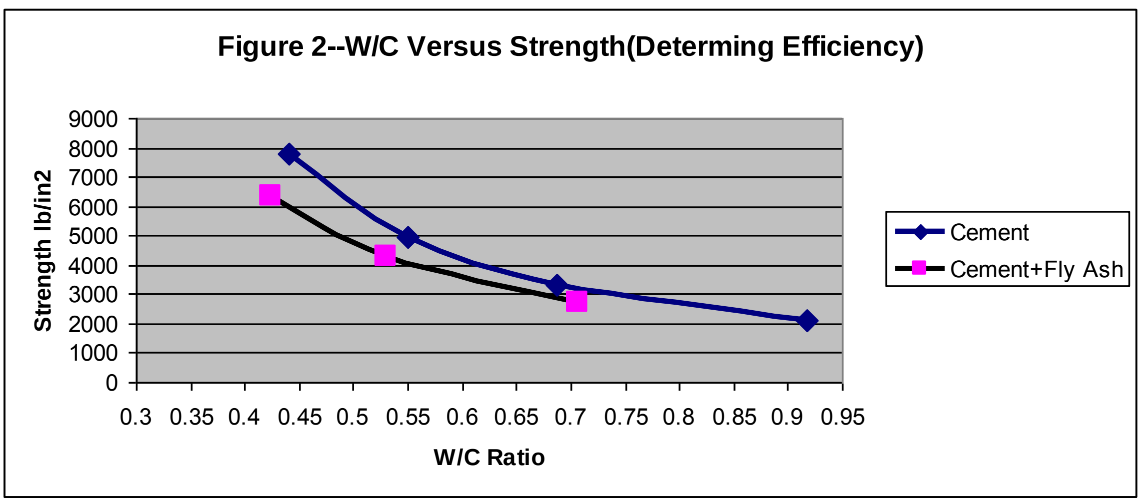
POZZOLANIC INDEX (How can it be determined?)

Pozzolanic index can be determined from two sets of concrete mixtures. In this discussion a 4-point curve (strength versus w/c water to cement ratio) for cement only mixtures and a 3 point curve (w/(c+f) water to cement plus fly ash ratio versus strength) for concretes containing 20% fly ash. Consider the concretes shown in Table 2 which have predicted model strengths as will be the strengths shown throughout the remainder of this paper.

Table 2—Concrete Mix Designs

Water (lb/yd^3)	Cement (lb/yd^3)	Fly Ash (lb/yd^3)	Predicted Strength (lb/in^2)
275	300		2140
275	400		3345
275	500		4950
275	625		7790
265	300	75	2710
265	400	100	4260
265	500	125	6355

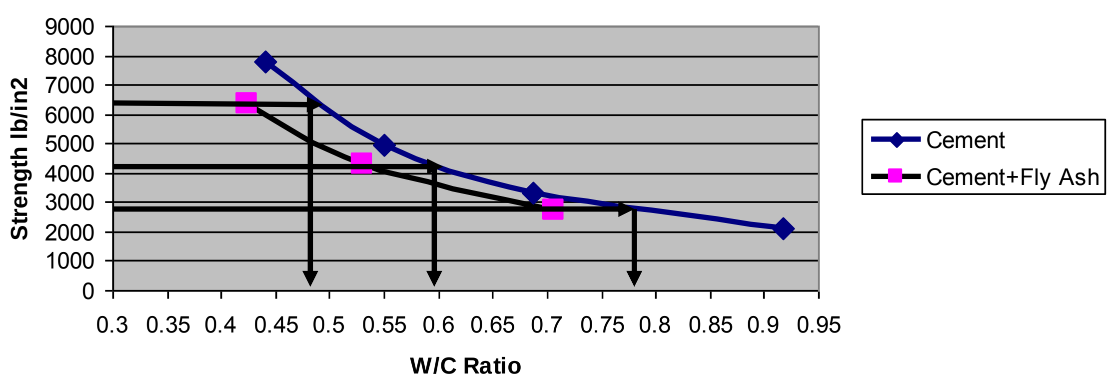
The results from Table 2 are plotted in Figure 2.



Fly ash efficiency can be determined using the values from Table 2 and from the curves shown in Figure 2 as illustrated in Figure 3. Horizontal lines (equal strength lines) are drawn through the fly ash strength points to the cement only curve. For each fly ash mix an equivalent cement only mix can be determined.

For the 400 cement and 100 fly ash concrete mixture, the equivalent cement only mix is at a w/c ratio approximately 0.595 just less than 0.60 as shown in Figure 3. The equivalent cement would be the water content of 275/0.595 = 462. lbs. The overall fly ash efficiency is then determined as (462-400)/100 = 0.62 (62%). The 100 pounds of fly ash is equivalent to 62 pounds more cement. This efficiency also includes the effect of water reduction. The water content of the cement only mixture is 275 lbs/yd^3 and 265 lbs/yd^3 for the fly ash mixture. Therefore the effect of chemical reaction (pozzolanic reaction) plus water reduction gives an overall efficiency of 62%. What is the chemical efficiency (pozzolanic reaction)? The equivalent cement with 265 water rather than 275 is is (462 * 265/275) = 445. The fly ash pozzolanic efficiency is then (445-400)/100 = 45%.

Figure 3--W/C Versus Strength(Determing Efficiency)



It is important to pick the cement only mixtures that extend beyond the mixtures with fly ash as shown in Figure 3, that is higher and lower strengths at the extremes.. In Figure 3, Table 2, the low end cement only mixture is 300 lbs of cement, which is the amount of cement in the lowest strength fly ash mixture, . The largest cement content for a cement only mixture is the same as the total cementitious materials in the highest strength mixture with fly ash, 500 lbs cement plus 125 lbs fly ash = 625 lbs of cement.

An alternate method to determine efficiency, rather than graphical, would be to use the information from the cement only mixtures and determine the empirical constants in Abrams' equation or another concrete strength equation. The w/c ratio of an equivalent cement mixture can then be determined by using the strength from a fly ash mixture in the Abrams strength equation or alternate equation. As in the graphical solution, the w/c is used to calculate the equivalent cement and thereafter the total and pozzolanic efficiency can be calculated.

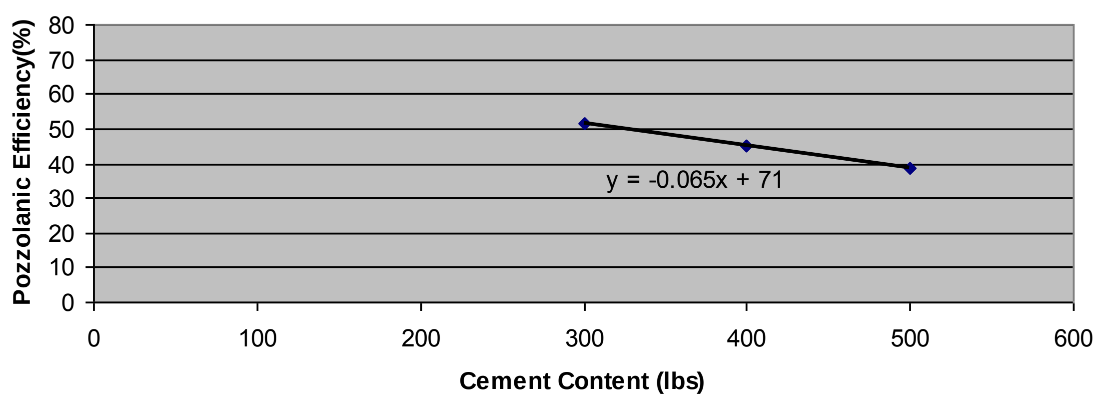
The overall total efficiency and pozzolanic efficiency by the graphical method for each fly ash concrete mixture is shown in Table 3.

Table 3—Fly Ash Efficiency

Concrete Mixture	Total Fly Ash Efficiency	Pozzolanic Efficiency
300 Cement+ 75 Fly Ash	69.0%	51.5%
400 Cement+100 Fly Ash	62.0%	45.0%
500 Cement+125 Fly Ash	54.0%	38.5%

A plot of cement content versus pozzolanic efficiency is shown in Figure 4. The y-intercept is “a” fly ash efficiency of 71.%. The efficiency of fly ash is directly proportional to the weight of pozzolanic reaction (equation 3 above 2.85 P_x P_w) specifically P_X. The y-axis of the figure is related to P_X from equation 3. The intercept of 70.833 is equal to 2.85 P_X therefore P_X = 71./2.85 = 24.91% approximately 25% (0.25). 25% of the fly ash has reacted. The pozzolanic reaction is 25% based on a silica equivalent. The mass of pozzolanic reaction is lime reaction with alumina, silica,. and possibly some iron of the fly ash and also includes alkali reactions with alumina, silica and iron. The 25% reaction in this discussion is then a silica equivalent.

Figure 4--Cement Content Versus Efficiency



The slope of -0.065 in Figure 4 is the relationship between cement reaction and pozzolanic reaction. The efficiency of cement changes with strength level. At lower water to cement ratios, cement efficiency is higher than at higher water to cement ratios. Thus if the cement efficiency is higher the pozzolanic efficiency (if it remains constant) would be lower as shown in Figure 4. Does P_X vary with w/c ratio? The author has found no information to suggest that pozzolanic reaction varies with w/c ratio. If pozzolanic index changed at the same rate as cement all of the efficiencies in Table 3 would be the same no matter the cement content. However, P_X may not be a constant. If pozzolanic reaction changes with heat, higher cement contents will generate more heat which could increase pozzolanic reaction. The author has not modeled a change in index due to increased internal heat.

CEMENT REDUCTION—POZZOLANIC REACTION

The amount of cement reduced was compared to pozzolanic reaction in Figure 3. The chemical pozzolanic efficiency is a constant for fly ash percentages $P_{\%}$ below the optimum. The optimum fly ash content is 35.1% from equation 5 [0.1352/(0.25 +0.1351)]. For example, with 15%, 25%, or 30% fly ash the pozzolanic efficiency would be the same as for 20% which is shown in Table 3 and Figure 5. The overall efficiency would vary depending how fly ash percentage affects water demand. The difference between overall efficiency and pozzolanic efficiency in Table 3 is the fly ash affect on water demand.

The maximum reduction in cement is at the optimum fly ash percentage which is approximately 35.1% as calculated above. Consider the following equations 6, 7 8 and 9.

$$C_R + P_E F = C_{initial} \qquad (6)$$

Where C_R is the new reduced cement content
P_E is the pozzolanic efficiency(decimal)
$C_{initial}$ is initial cement content(no pozzolan)

$$F = 0.351\, C_R/(1-0.351) \qquad (7)$$

Fly ash content at optimum
Optimum in this paper for P_X = 0.25 is 35.1%(0.351)
Equation 5

Substituting equation 6 for F in equation 5 gives the following:

$$C_R = C_{initial} / (1 + 0.5408\, P_E) \qquad (8)$$

The amount of cement is also reduced by the amount of pozzolan(fly ash) water reduction as shown in equation 8:

$$C_{RW} = C_R W_R / W_{initial} \qquad (9)$$

Where C_{RW} is the reduced cement content due to pozzolanic reaction and water reduction
W_R is the reduced water content
$W_{initial}$ is the initial water content

For 500 lbs of cement the pozzolanic efficiency is 38.5% (0.385) From equation 7 the reduced cement content C_R = 500/(1 + (0.5408 *0.385)) = 414 lbs. The reduced water content based on 35.1% fly ash and as estimated in this paper is 257.5 lbs and the initial water is 275 lbs Using equation 8, C_{RW} = 257.5 * 414/275 = 387 lbs. This is a cement reduction of 500-387 = 22..6%. With 387 lbs of cement and an optimum fly ash percentage of 35.1% the fly ash content can be calculated by equation 6 replacing C_R with C_{RW}. The fly ash content is (388 * 0.351/(1-.351) = 210 lbs. Similar calculations for 300 and 400 lbs of cement are shown in Table 4.

Table 4—Concrete Mixtures at Optimum Fly Ash Percentage

Initial Cement-lbs	Water-lbs	Cement-lbs	Fly Ash-lbs	Cement Reduction %	Predicted Strength Lbs/in^2
300	257.5	220	118	26.8	2160
400	257.5	302	163	24.5	3400
500	257.5	387	210	22.6	4995

The predicted strengths shown in Table 4 are comparable to the cement only strengths for 300, 400 and 500 lbs of cement in Table 2.

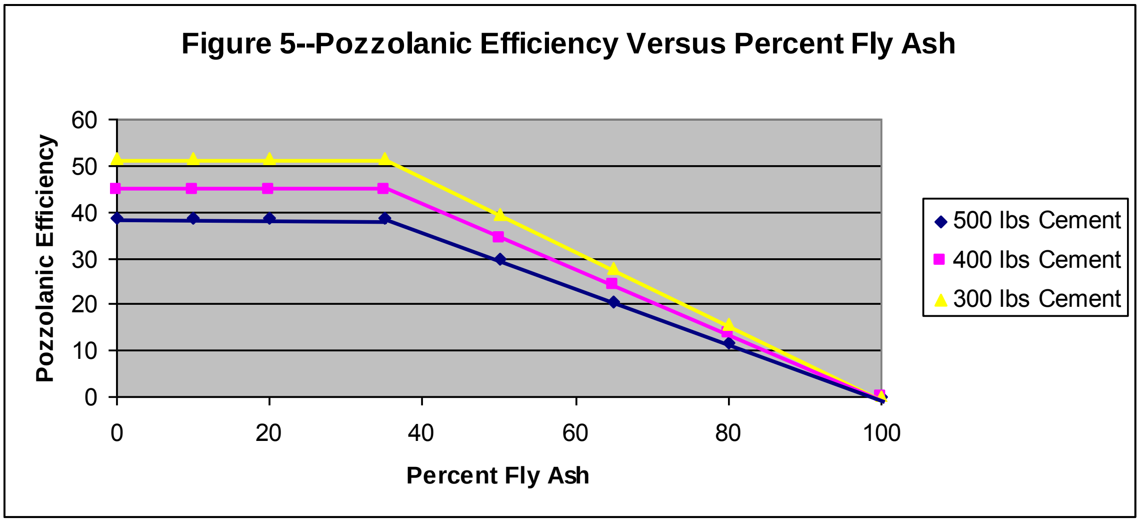
CEMENT REDUCTION—POZZOLAN PERCENTAGE ABOVE OPTIMUM

When the percentage of fly ash is greater than optimum the pozzolanic efficiency will be reduced. For concretes with percentages greater than optimum there will not be enough lime to react with the reactive portion of the fly ash. The mass of pozzolanic reaction is the following for percentages greater than optimum:

$$((H_X C_W)/3)*1.5405 \qquad (6)$$

Where H_X is the amount of cement that has reacted
C_W is the mass of cement in the concrete
1.5405 = 342/222 from equation 1.
“3”--The lime produced is 1/3 of reaction products of cement

Chemical pozzolanic efficiency is illustrated in Figure 5 for P_X = 0.25 with the optimum fly ash percentage at 35.1%.



As illustrated in Figure 5, pozzolanic efficiency is related to cement content and for each content, efficiency is a constant for fly ash percentages less than optimum. The optimum is assumed to be the point at which all of the lime is consumed by the reactive portion of the fly ash. At fly ash percentages greater than the optimum there is less lime available (less cement to produce lime) than at optimum therefore the pozzolanic efficiency is reduced down to 0 at 100% fly ash and no cement(no lime).

CEMENT REDUCTION—LEED Green Concrete

For a special LEED credit the cement content is reduced by 40%. As shown in Table 4 pozzolanic efficiency and pozzolanic (fly ash) water reduction can not reduce cement content by 40%. Reductions in cement in Table 4 range from 22.6% to 26.8%. Pozzolanic reaction and pozzolanic (fly ash) water reduction cannot produce a 40% reduction in cement. Additional water reduction is needed. A higher percentage of pozzolan will be of no benefit with pozzolanic efficiency but may produce additional water reduction.

As shown in Table 2 fly ash reduces water content. The reduction in this paper is for each 20% of fly ash the water is reduced by 10 pounds. The fly ash percentages in Table 2 are 20% so the water reduction is 10 lbs from 275 cement only to 265 for concrete mixtures with fly ash. This reduction will vary with each set of materials, cement, fly ash and aggregates. It might be much more or much less. Using the water content relationship in Table 2 and the efficiencies in Table 3 is it possible to reduce the cement content by 40% for a special LEED credit. Let's begin with the 500 lbs cement mixture in Table 2 last mixture Table 5. The water to cement ratio is 275/500 = 0.55. A reduction of 40% would reduce the cement content to 300 lbs. For 300 lbs of cement the pozzolanic efficiency is 51.5% (Table 3). The optimum fly ash percentage is 35.1% which is [300/(1-0.351)]*0..351 = 162 lbs of fly ash. The water reduction is estimated to be 10*35/20 = 17.5 lbs which is 275-17.5 = 257.5. The effective cement at optimum is 300 lbs of cement + (162 * 0.515) = 383 lbs (effective cement) and the water content is estimated to be 257.5 lbs with an estimated strength of 4605 lb/in^2 which is lower than the desired strength of 4950 lbs/in^2. To obtain the desired strength the water content must be lowered by the use of a high range water reducer. The comparison strength is for a mixture with 500 lbs of cement. The estimated water required water (383/500) * 257.5.= 197 lbs/yd^3. The reduction from 257.5 to 197 can be obtained by using a high range water reducer. This is a reduction of 257.5-197 = 60.5 lbs, or 60.5/257.5 = 24.4%. High range water reducers can potentially reduce water content by 25% or more.

The water reduction could be a combination of fly ash and high range. A common LEED mix design is 50% fly ash. This would be a mix with 250 lbs of water 300 lbs of cement and 300 lbs of fly ash with an estimated strength of 3535 lb/in^2. To obtain a water content of 197 lbs this is a water reduction of (250-197)/250 = 21.2% which would be easier to achieve with a high range water reducer than for the mixture at optimum discussed above.

The mix designs described in this "LEED Green Concrete" section are shown in Table 5.

Table 5—LEED Concretes

Water Lbs/yd^3	Cement Lbs/yd^3	Fly Ash Lbs/yd^3	Predicted Strength Lbs/in^2 28-days
275	300		2140
257.5	300	162	3365
197	300	162	5100
250	300	300	3535
197	300	300	5100
275	500		4950

For comparison to real life concretes, the 197 lbs of water, 300 lbs of cement and 300 lbs of fly ash, (197/(300+300))=0.33 water to cement ratio concrete is common for 4000 lb/in^2 design strength (performance 5000 lb/in^2).for mixtures used in the San Francisco Bay area for LEED concretes[7].

GREEN CONCRETE

The National Ready Mixed Concrete Association has recently developed a "Sustainable Concrete Carbon Calculator". As expected the primary source of CO_2 (carbon) is Portland cement. More than 90% of the calculated carbon is due to cement. Therefore reducing cement is primary in making concrete more "green". This calculator shows for a 12% reduction in cement the carbon is reduced by 11%, a reduction of 23% reduces carbon by 21% and a reduction in cement of 40% reduces carbon by 37%. There is no real difference in carbon when comparing the 300 cement and 162 fly ash mixture to the 300 cement and

300 fly ash mixture shown in Table 5. However the lower fly ash content mixture would be more economical to produce.

The higher fly ash content mixture (50%) would have much more non-hydrated fly ash which could potentially cause dusting or surface weakness and thus be less durable.

The amount of cement reduction is a measure of “green” however the percentage of fly ash itself is a misleading indicator of “green”.

CAVEATS

1. The concrete mixtures in this paper and strengths were developed from a concrete strength model. The model gives excellent predictions on strengths when compared to historical Florida Concretes. However, there may be unknown factors that have not been considered that would produce some variation in real world trial batches when compared to the strengths predicted in this paper.

2. The amount of water reduction due to fly ash may be much higher or lower than that assumed in this paper which would significantly change the amounts of cement and fly ash for the mixtures described in the paper.

3. Pozzolanic reaction is not completely a silica reaction. The constant 2.85 used in paper could be higher. For alumina reactions it would be 3.18. The real number likely varies for each fly ash. True pozzolanic reaction may not be exactly those shown (equations 1 and 2). A “constant” different than 2.85 would produce a different pozzolanic index and a different corresponding optimum fly ash percentage.

4. The amount of water consumed by pozzolanic reaction (equation 1) is only that in the free lime. Equation 2 indicates that alumina reactions consume additional water. What impact does this have on cement hydration, if any? A change in cement hydration relative to pozzolanic reaction would change the efficiencies calculated. A change in cement hydration patterns due to fly ash chemical reaction is not included in the model used in this paper.

5. The model and methods used in this paper work for materials that are predominately pozzolanic such as low CaO content Class F Fly Ashes, Silica Fume, and Calcined Clay. The methods in this paper will not work for Class C Fly Ashes that are both pozzolanic and cementitious and it does not work for blast furnance slags that are predominately latently hydraulic and pozzolanic.

CONCLUSIONS

1. Pozzolanic reaction can significantly reduce the required amount of cement and make concrete “green”. The greatest reduction due to pozzolanic reaction is found to be at the optimum percentage.

2. Fly Ash (Pozzolan) can also make concrete “green” by reducing the water content which in turn reduces cement content. The amount of water reduction in conjunction with pozzolanic reaction may not be enough to reduce cement by more than 25%, therefore for a 40% reduction in cement the use of chemical admixtures such as high range water reducers may be required.

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