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Guide to Using HYDRA3D: A Three-Dimensional Digital-Image-Based Cement Microstructural Model

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ABSTRACT

A computer program, HYDRA3D, to simulate cement microstructural development and quantify microstructural characteristics has been developed. HYDRA3D is a menu driven program available in either Fortran or C which allows a user to create a starting microstructure, execute hydration, measure phase fractions, and assess phase connectivity. This manual outlines the conceptual model on which HYDRA3D is based, describes the programs in detail, and provides examples of program usage. A system calling diagram, source code listings, guidelines for modifying the programs, and system requirements are provided in the Appendices.

Keywords: Cement, computer modelling, hydration, interfacial zone, microstructure, percolation, simulation, software.

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1. INTRODUCTION

In the past few years, researchers at the National Institute of Standards and Technology (NIST) have developed a three-dimensional digital-image-based model for simulating the microstructural development of hydrating cement (specifically tricalcium silicate, C_3S[1]) paste [1-3]. The advantages of a digital-image-based approach over a continuum approach are several. First, since each particle in the model is a collection of pixels (or voxels), real particle shapes may be used as well as conventional geometric shapes such as spheres. Second, the rules for microstructural development can be mechanism-based, as the processes of diffusion, nucleation, and surface reaction can be conveniently simulated on a lattice of pixels. Third, since the microstructure is defined on a lattice, techniques may be borrowed from statistical physics to easily assess material properties such as percolation [4], electrical conductivity [5], and ionic diffusivity [6]. Finally, by utilizing a graphics workstation, the simulated microstructural development can be visualized "in situ" to provide new insights into the hydration process and the relationships between microstructure and material properties.

Over time, interest in the model by the cement research community outside of NIST has increased to a point where other researchers have requested copies of the model computer program, **HYDRA3D**. It is intended that this document serve as a technical guide to using HYDRA3D as well as providing sufficient details that the interested user might modify the code to better serve their purpose. The model code is available from the Cementitious Materials Modelling Laboratory at NIST. The authors may be contacted for details concerning acquisition.

Section 2 of this manual provides an overview of the conceptual model on which HYDRA3D is based. Section 3 describes the programs (HYDRA3D.F and HYDRA3D.C as the code has been developed in both Fortran and C), detailing both the menu system by which the user controls program execution and the separate modules (subroutines) comprising the overall code. Example runs including results are provided in section 4. A calling diagram and commented source code listings are given in the Appendices along with suggested guidelines for modifying the source code and information on system requirements for executing the model.

2. MODEL DESCRIPTION

The key to the microstructural model is the representation of a unit volume as a discrete three-dimensional array of elements (pixels). Each element is uniquely identified as belonging to a single phase of the hydrating cement system and is

[1] Standard cement chemistry notation is used throughout this document. That is, $C=CaO$, $S=SiO_2$, $H=H_2O$, $A=Al_2O_3$, and $F=Fe_2O_3$.

typically 1 μm on a side. The hydration process is simulated by performing operations on these individual elements according to a prescribed set of rules as outlined below.

A starting cement microstructure consists of digitized spherical particles randomly dispersed in water. Thus, each C_3S particle consists of a set of pixels representing a sphere of a desired diameter (e.g. 3 to 20 μm). The spheres are placed at random (x,y,z) locations in the volume such that they do not overlap one another. Since the model is relatively small, being 100*100*100 pixels in size, periodic boundaries are utilized to eliminate artificial edge effects. Thus, if a spherical particle extends out one face of the volume, it is completed on the opposite side. The cement particles are added in order of largest to smallest to aid in achieving a random dispersion without having to relocate the smaller particles to fit the larger ones into the system.

In addition to cement particles, two other types of particles may be added to the system. First, a single flat aggregate of user-specified thickness may be placed in the middle of the 3-D system. The aggregate extends the length of the system in the y and z directions. This allows one to study the microstructural development occurring in the interfacial zone in a model concrete system [7,8]. Second, inert or pozzolanic mineral admixture particles may be added to the system. In the case of pozzolanic particles, the particles react with the CH produced during hydration to produce pozzolanic or secondary C-S-H. Pozzolanic admixture particles are assumed to be pure silica with a molar volume of 27 cm^3/mole but these assumptions may be modified as outlined in Appendix D. The mineral admixture particles may be either 1 pixel (μm) in size or of some variable diameters as in the case of the C_3S particles.

The user may control the water-to-cement (w/c) ratio of a starting microstructure by varying the number and size of C_3S particles added to the system. In a system containing only cement and water, if f is the volume fraction of pixels which are C_3S, then

$$\frac{w}{c}=\frac{1-f}{3.2 * f} \qquad (1)$$

where 3.2 is the specific gravity of cement. For systems containing aggregates and/or mineral admixtures, the above equation no longer holds. In such systems, to determine the water-to-solids (w/s) ratio, the general equation

$$\frac{w}{s}=\frac{1-f_{agg}-f_{cement}-f_{\text{min. adm.}}}{\rho_{cement} * f_{cement}+\rho_{\text{min. adm.}} * f_{\text{min. adm.}}} \qquad (2)$$

where f_i and ρ_i represent the volume fraction and density of component i respectively, should be utilized.

The user of the model should keep in mind that the mineral admixture and cement are typically of different densities, so that weight and volume fractions are not identical. For example, the density of cement is about 3.2 g/cm^3 while that of silica fume is typically 2.2 g/cm^3. Thus, replacing 10% by weight of the cement with silica fume results in a system in which 13.9% of the total solids volume is silica fume.

Hydration of a starting microstructure is modeled as an iterative process consisting of discrete cycles as illustrated schematically for a two-dimensional system in Figure 1. Each cycle consists of three processes: dissolution, diffusion, and reaction. Basically, material dissolves off the surfaces of the original cement particles, diffuses around within the available pore space, and reacts to form hydration products. The reactions are based on the hydration of tricalcium silicate which is assumed to react with water as follows [9]:

$$C_3S + 5.3H \rightarrow C_{1.7}SH_{4.0} + 1.3CH \quad (3) \ .$$

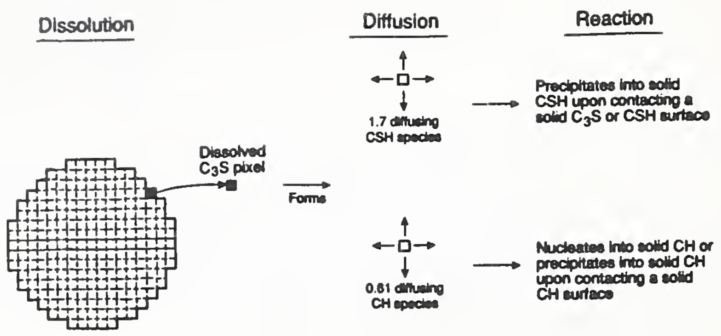
On a volume basis, this reaction is equivalent to 1 unit of C_3S producing 1.7 units of C-S-H and 0.61 units of CH [9]. Thus, 1 unit of solid reactant reacts with water to produce 2.31 units of solid product. This volume expansion in terms of solids is responsible for the evolution of a cement paste from a colloidal dispersion into a rigid solid material. The similarity between this expansion factor for C_3S hydration and cement (C_3S, C_2S, C_3A, C_4AF, and gypsum) hydration has been discussed elsewhere [4,6] and is one basis for considering the model to be a true model of cement microstructure and not limited solely to pure C_3S. Indeed, favorable comparisons between model and real cement systems have been made [1,4-8].

When pozzolanic mineral admixtures are present in the system, diffusing CH species are allowed to react at pozzolanic surfaces according to:

$$1.7\ CH + S + 2.3\ H \rightarrow C_{1.7}SH_{4.0} \quad (4) \ .$$

Assuming specific volumes of 27 cm^3/mole for S, 33.1 cm^3/mole for CH, and 124 cm^3/mole for C-S-H, this reaction is equivalent to one unit of mineral admixture reacting with 2.08 volume units of CH to produce 4.6 volume units of pozzolanic or secondary C-S-H. Conversely, inert mineral admixtures do not react with any of the cementitious species present in the system.

In the dissolution phase, the entire 3-D microstructure is first scanned to identify all C_3S pixels which have one or more neighbors which are water-filled porosity. Six neighbors (± 1 in the x, y, and z directions) are checked to evaluate this criteria. Next, in a second pass through the microstructure, each C_3S pixel identified in the first pass attempts to take a one step random walk. If the step is into porosity, the C_3S pixel is dissolved and a diffusing C-S-H species is created at the step's destination location. If the step is into solid material, the C_3S pixel remains as solid



Dissolution
Diffusion
Reaction
1.7 diffusing CSH species
Precipitates into solid CSH upon contacting a solid C_3S or CSH surface
Dissolved C_3S pixel
Forms
0.61 diffusing CH species
Nucleates into solid CH or precipitates into solid CH upon contacting a solid CH surface

Figure 1. Schematic Diagram of Steps in the Hydration Model

C_3S for this cycle of the hydration. This algorithm produces the desired effect that sharp edges will dissolve at a faster rate than flat surfaces and smaller particles will dissolve at a faster rate than larger ones due to their larger surface to volume ratio. Finally, after all C_3S pixels have been checked for dissolution, extra CH and C-S-H diffusing species are added at random locations in the water-filled space to maintain the correct volume stoichiometry according to equation 3. For example, if in a given cycle, 100 pixels of C_3S dissolve, 70 extra pixels of diffusing C-S-H and 61 extra pixels of diffusing CH would be added to the system.

Once the correct number of diffusing species have been created, each one executes a random walk in the available porosity until reaction occurs. Thus, for each diffusion step, every diffusing species remaining in the system continues its random walk. For each step, a direction is chosen at random and the diffusing species is allowed to move one pixel in the chosen direction if that pixel is currently unoccupied (porosity). Since diffusing species may occasionally become trapped in regions where no reaction (based on the rules outlined below) may occur, after some large number of steps, all remaining diffusing species are converted into solid product.

The reaction rules differ for the C-S-H and CH diffusing species. The C-S-H species execute random walks in the pore space until they encounter (run into) a solid C_3S or solid C-S-H surface. When this event occurs, the diffusing C-S-H species is converted into solid C-S-H. These rules for C-S-H formation combine aspects of both the through solution (diffusion) and topochemical mechanisms (surface reaction) for C-S-H formation outlined in the cement literature [10].

Solid CH forms by a nucleation and growth process in the pore space. For each random step taken by a diffusing CH species, there is a probability that it will nucleate at its current location. This probability is exponentially proportional to the number of diffusing CH species remaining in pore solution and is given by

$$P_{nuc}=P_0 * [1.- \exp(\frac{-[CH]}{[CH]_{\max}})] \quad (5)$$

where P_{nuc} is the probability of nucleation, P_0 is the maximum probability of nucleation, [CH] is the number of CH diffusing pixels remaining in solution at each step, and $[CH]_{max}$ is a scale factor [7]. By varying P_0 and $[CH]_{max}$, the number and size of CH crystals formed during the hydration can be controlled. In addition to nucleation, if a diffusing CH species encounters a solid CH surface during its random walk, it is converted into solid CH at its present location, resulting in growth of the CH crystal.

When pozzolanic mineral admixtures are present in the system, the diffusing CH species may react at the pozzolanic surfaces to form pozzolanic C-S-H. Since according to equation 4, this reaction is expansive in terms of solids, there is a probability that two volume elements of pozzolanic C-S-H instead of one are formed whenever this reaction occurs. This probability is 0.73 as 2.08 units of diffusing CH should produce 3.6 units of pozzolanic C-S-H, in addition to the volume element originally occupied by the pozzolanic mineral admixture. This extra pozzolanic C-S-H is added at a random neighboring location if possible, and at a random location in the pore space otherwise. When all of the pozzolanic mineral admixture has been consumed by reaction with diffusing CH, the pozzolanic reaction is discontinued.

When all diffusing species generated during a given dissolution phase have reacted, a new hydration cycle is begun with a new dissolution. The degree of hydration, α, after m cycles of hydration is given by

$$\alpha(m)=\frac{[C_3S]_0-[C_3S]_m}{[C_3S]_0} \quad (6)$$

where $[C_3S]_i$ is the number of solid C_3S elements remaining after i cycles of hydration. Within HYDRA3D, the user may specify either the number of hydration cycles to execute or the desired degree of hydration to be achieved.

Since the model microstructure is available in a digitized format, phase fractions can be easily assessed by simply counting the number of pixels of each phase. These phase fractions may be assessed globally or as a function of distance from the aggregate surface when an aggregate is present in the system. In addition, due to the underlying 3-D lattice structure of the model, the connectivity or percolation of the individual phases or total solids may be easily determined using a simple burning algorithm [11]. This algorithm is a simple way of identifying all pixels that are part of a spanning cluster, if such a cluster exists, and works as follows. Conceptually, all the pixels belonging to the phase(s) of interest are classified to be "combustible". A "fire" is started on one side of the model's unit cell, and allowed to propagate only along these combustible pixels. Within HYDRA3D, this burning algorithm is executed in a non-periodic manner such that the fire cannot exit one side of the unit cell and enter the other (like the diffusing species can during the hydration). If any pixels on the opposite side of the model cell are found to have been "burned", then a spanning cluster of the phase of interest must exist. The number of "burned" pixels are counted to determine the fraction of the phase of interest that is a part of the spanning cluster. This connectivity can be assessed after any number of hydration cycles.

3. PROGRAM DESCRIPTION

In developing HYDRA3D, an effort has been made to incorporate the guidelines for the development of computer-based models as outlined by Kaetzel et al [12]. Three major software engineering considerations for such code are accessibility, maintainability, and transportability. To increase accessibility, the code for the digital-image-based hydration model has been developed in both Fortran and C, enabling transfer to a wider audience of researchers. Additionally, a menu system has been incorporated into the programs to guide the user in executing the code and to provide flexibility in the analyses selected by the user. A modular approach, as shown in the calling diagram provided in Appendix A, has been utilized to enhance code maintainability. Transportability has been enhanced by including a portable random number generator [13] as one of the modules of the code. It is hoped that these steps will increase the usability of HYDRA3D by other cement researchers.

3.1 MENU SYSTEM

The program is menu driven, with the user being prompted to supply input parameters based on their selection from the main menu shown in Figure 2. Each menu choice is described in detail below.

```
Input User Choice:
1) Add a flat aggregate to microstructure
2) Add spherical particles (C3S or filler) to microstructure
3) Add one-pixel filler particles to microstructure
4) Hydrate microstructure
5) Measure phase fractions
6) Measure phase fractions as a function of distance from
   aggregate surface
7) Measure single phase connectivity
8) Measure total solids connectivity
9) Exit
```

Figure 2. Main Menu for HYDRA3D

1) Add a flat aggregate to microstructure

This selection is utilized to place a thin plate-like aggregate in the middle of the 3-D microstructure. The aggregate extends the length of the box (100 pixels) in the y and z directions with the user specifying the thickness in pixels in the x direction. The input thickness should be an even integer to maintain system symmetry, with equal numbers of (paste) pixels to the left and right of the centered aggregate. Under normal operating conditions, if an aggregate is desired, it should be added to the system before any cement or mineral admixture particles. If any such particles are in the system when the aggregate is placed, it will be superimposed over those particles with all pixels within the aggregate boundaries being set to the phase ID of aggregate.

2) Add spherical particles (C_3S or filler) to microstructure

Via this selection, the user may add spherical (digitized spheres) cement or mineral admixture particles to the 3-D microstructure. Once selected, the user will first be prompted to enter the number of different sizes of spheres to be placed. Next, the user will be prompted to enter the number, radius, and phase ID of the spheres for each size class. The user should begin with the largest radius size class and proceed sequentially to the smallest radius size class. The specified spheres will be placed at random locations in the 3-D microstructure using periodic boundaries as described in section 2.

When a user is adding both mineral admixture and C_3S particles of the same radius, they should place all the largest particles (C_3S and filler) first, all the 2nd largest particles next, etc. This provides the best chance for all particles to be placed. If some of the smaller particles are placed before the larger ones, there may be no remaining spaces into which the larger ones can fit. This procedure is illustrated in one of the examples given in section 4.2.

3) Add one-pixel filler particles to microstructure

This selection is used to add small (1 pixel or 1 μm) mineral admixture particles to a starting microstructure. The user must supply the number of particles to be added and whether they are inert or pozzolanic. Since these particles are typically smaller than the cement particles used in the model (just as silica fume is finer than cement), they should be added after the cement particles and larger filler particles (if any) have been placed in the system.

4) Hydrate microstructure

This selection is utilized to execute the hydration model on a 3-D microstructure. As described in section 2, a given cycle of hydration consists of dissolution, diffusion, and reaction. When this menu item is selected, the user will be prompted to choose either to specify the number of hydration cycles to execute or the desired degree of hydration and must supply a numerical value for the item selected. Additionally, the user must specify the maximum number of diffusion steps to take in a cycle before converting all remaining diffusing species to product (see section 2), and two parameters to control CH nucleation based on equation 5. The value for the maximum number of diffusion steps to execute in a given cycle is generally on the order of 5000 to 10000. Typical values for the two parameters controlling CH nucleation would be 0.001 for the maximum probability of CH nucleation, P_0, and 200000. for the exponential scale factor, $[CH]_{max}$.

5) Measure phase fractions

This selection will simply return counts of the volume (pixels) occupied by each phase in the microstructure. This item is useful in verifying the original w/c ratio of the system and to determine the degree of hydration, based on the C_3S remaining after some number of cycles of hydration.

6) Measure phase fractions as a function of distance from the aggregate surface

When an aggregate is present in the 3-D microstructure, this selection may be utilized to measure the phase fractions present as a function of distance from the surface of the aggregate. Output consists of a table of the phase fractions of all phases present for each (pixel) distance from the aggregate. This selection allows one to study the microstructure of an interfacial zone in a cement-based composite [7,8].

7) Measure single phase connectivity

This selection allows the user to determine the percolation characteristics of any individual phase (porosity, etc.) present in the microstructure. The user will be prompted to input the ID of the phase of interest, after which the program will determine the number of pixels of this phase which are accessible from the top of the system and the number of pixels of this phase which are part of a pathway(s) through the system going from top to bottom. The burning algorithm described in the end of section 2 is utilized to determine these values.

8) Measure total solids connectivity

This selection is identical to selection #7 except that the percolation characteristics of all solids present in the system (with the exception of any aggregate) are assessed. As the hydration proceeds, the solids will eventually form a rigid connected structure which spans the 3-D microstructure. This point should correspond to the set point (time) commonly measured by cement researchers.

3.2 MODULE DESCRIPTIONS

Program Name: MAIN
Purpose: To initialize 3-D microstructure, present the user with a menu of choices, and call routines to execute selected menu options
Calls: INTRO, ADDAGG, CREATE, FILLER, HYDRATE, MEASURE, MEASAGG, CONNECT, CONSOLD
I/O: Outputs menu of selections to user
User inputs integer seed for random number generator
User inputs integers corresponding to menu choices

Module Name: INTRO
Purpose: To display an introduction to HYDRA3D to the system user
Called By: Main program
I/O: Introductory text is output to the standard output unit (screen)

Module Name: ADDAGG
Purpose: To place a single plate-like aggregate in the middle of the 3-D microstructure.
Called By: Main program
I/O: User inputs an even integer for aggregate thickness
Notes: Aggregate extends length of the system in the y and z directions and specified thickness in x direction.
Aggregate is assigned a phase ID of 8 and is inert with respect to diffusing C-S-H and CH species.

Module Name: CREATE
Purpose: To obtain user input specifying spherical particles to be placed in 3-D microstructure and call routine to execute placement.
Called By: Main program
Calls: GSPHERE
I/O: User inputs parameters specifying spherical particles to be generated
Number of classes
Number of spheres, radius, and phase ID for each class

Module Name: GSPHERE
Purpose: To generate digitized spheres of a fixed number of classes by finding random (x,y,z) locations at which the spheres can be placed.
Called By: CREATE
Calls: CHKSPH, RAN1
Passed In: Number of classes to be generated
Arrays of number, radius, and phase ID for each class
Notes: For each random location generated, sphere placement is first tested (no overlap with existing spheres) and then, if possible, performed.

Module Name: CHKSPH
Purpose: To check or perform placement of a digitized sphere of specified radius and center location by operating on all pixels falling within the boundaries of the sphere.
Called By: GSPHERE
Passed In: X, Y, and Z coordinates for sphere center
Radius of sphere
Flag indicating if sphere placement is to be checked or performed
Returns: Flag indicating if sphere placement is possible
Notes: Periodic boundaries are employed in checking and placing spheres.
Sphere diameter = 2*sphere radius + 1 so that spheres can always be centered on a pixel.

Module Name: FILLER
Purpose: To obtain user input as to the number and type of filler particles to be generated and call routine to execute placement.
Called By: Main program
Calls: GENFILL
I/O: User inputs number and phase ID of filler particles to be generated

Module Name: GENFILL
Purpose: To place one pixel mineral admixture particles at random unoccupied (pore) locations in 3-D microstructure.
Called By: FILLER
Calls: RAN1
Passed In: Number and phase ID of 1-pixel filler particles to generate

Module Name: HYDRATE
Purpose: To obtain user input and control hydration for some number of cycles or to some degree of hydration.
Called By: Main program
Calls: DISSOLV, MVCHANT, MCSHANT
I/O: User inputs parameters to control hydration
Number of hydration cycles or degree of hydration to execute
Maximum number of diffusion steps per cycle
Maximum probability for CH nucleation
Exponential scale factor for CH nucleation
Outputs number of diffusing species generated during each hydration cycle
Notes: For each cycle, hydration is controlled by calling a routine to perform the dissolution and then calling routines to move the diffusing C-S-H and CH species until all diffusing species have reacted.

Module Name: DISSOLV
Purpose: To perform dissolution for each hydration cycle.
Called By: HYDRATE
Calls: RAN1
Returns: Arrays holding x, y, and z locations of generated diffusing species and number of diffusing species generated.
Notes: Dissolution is performed by randomly dissolving C_3S species in contact with pore space and creating the appropriate numbers of diffusing C-S-H and CH species.
Extra diffusing species are added at totally random unoccupied locations in the 3-D microstructure.

Module Name: MVCHANT
Purpose: To execute a single diffusion step for a diffusing CH species.
Called By: HYDRATE
Calls: RAN1, ADDEXT
Passed In: Current location of diffusing CH species
Current nucleation probability for CH
Returns: New location of diffusing CH species
Flag indicating if reaction has occurred
Notes: If nucleation is probable, the diffusing CH species is converted to solid CH at its current location. Otherwise, the diffusing CH species undergoes a one-step random walk in 3-D. If it collides with solid CH, it is converted to solid CH. If it collides with pozzolanic material, the pozzolanic reaction occurs as long as sufficient pozzolanic mineral admixture remains in the system. If the step is into pore space, the location of the diffusing CH species is updated and returned to the calling routine. If any reaction occurs, a flag is set and also returned to the calling routine.

Module Name: ADDEXT
Purpose: To add extra pozzolanic C-S-H to microstructure when a diffusing CH species reacts at a pozzolanic surface.
Called By: MVCHANT
Calls: RAN1
Passed In: X, Y, and Z coordinates of reaction site
Notes: The six neighboring locations of the reaction site are sampled at random, looking for an unoccupied site to place the extra pozzolanic C-S-H. If no such site exists, the extra pozzolanic C-S-H is placed at a random unoccupied location in the 3-D microstructure.

Module Name: MCSHANT
Purpose: To execute a single diffusion step for a diffusing C-S-H species.
Called By: HYDRATE

Calls: RAN1
Passed In: Current location of diffusing C-S-H species
Returns: New location of diffusing C-S-H species
Flag indicating if reaction has occurred
Notes: The diffusing C-S-H species executes a one-step random walk in 3-D. If it collides with solid C-S-H or C_3S, it is converted to solid C-S-H. If the step is into pore space, the location of the diffusing C-S-H species is updated and returned to the calling routine. If reaction occurs, a flag is set and also returned to the calling routine.

Module Name: RAN1
Purpose: To generate random numbers for use by other routines in the model program.
Called By: GSPHERE, GENFILL, DISSOLV, MVCHANT, ADDEXT, MCSHANT
Passed In: Seed for random number generator
Returns: A real number in the range [0.0, 1.0)

Module Name: MEASURE
Purpose: To determine phase fractions of all phases present in the 3-D microstructure.
Called By: Main program
I/O: Outputs global phase counts (in pixels) to the standard output unit (screen)

Module Name: MEASAGG
Purpose: To assess phase fractions as a function of distance from aggregate surface to enable quantitative characterization of interfacial zone microstructure.
Called By: Main program
I/O: Outputs phase fractions as a function of distance from aggregate surface in tabular format

Module Name: CONNECT
Purpose: To assess the percolation characteristics of an individual phase using a simple burning algorithm.
Called By: Main program
I/O: User inputs ID of phase of interest
Outputs number of elements of phase which are accessible from top
Outputs number of elements of phase which are part of pathways through the 3-D microstructure from top to bottom
Notes: Burning algorithm is non-periodic.

Module Name: CONSOLD
Purpose: To assess the percolation characteristics of total solids present in the 3-D microstructure using a simple burning algorithm.

Called By: Main program
I/O: Outputs number of elements of total solids which are accessible from top
  Outputs number of elements of total solids which are part of pathways through the 3-D microstructure from top to bottom
Notes: Aggregate is not included in total solids as it always forms a continuous path from top to bottom of the 3-D microstructure.
  Burning is non-periodic.

4. EXAMPLES

HYDRA3D can be executed either interactively or in a batch mode by piping the standard input and output to datafiles. On a UNIX-based system, for example, typing

```
hydra3d <hydrate.inp >hydrate.out
```

would execute the program hydra3d, reading the menu selections and other input from the datafile hydrate.inp and outputting all results to hydrate.out. The output file could then be imported to a spreadsheet or graphical analysis package for further analysis as illustrated below. All examples were executed in this batch mode using the Fortran version of HYDRA3D.

4.1: EFFECT OF MINERAL ADMIXTURE PARTICLE SIZE ON INTERFACIAL ZONE MICROSTRUCTURE

For this example, we will create starting systems consisting of an aggregate, cement particles, and either large or one-pixel mineral admixture particles added at 20% replacement for cement on a weight basis. The one pixel admixture particles would represent well dispersed silica fume while the larger admixture particles would be indicative of an agglomerated silica fume system. The systems will be hydrated to the same degree of hydration and the interfacial zone microstructures compared based on the phase fractions present as a function of distance from the aggregate surface. A w/s ratio of 0.45 will be utilized and hydration will be executed to 70%. The cement particles (and large mineral admixture particles) will consist of equal volume fractions of particles 21 and 11 pixels in diameter while the aggregate thickness will be set at 2 pixels. The user of the system must decide on these system variables before a simulation can be executed.

To know how many of each size particle are needed, the user must know how many pixels are occupied by a single sphere of each particle radius. These values are tabulated in Table I for sphere radii ranging from 1 to 10. As noted earlier, the sphere diameter is equal to 2* radius +1 so that the spheres may always be centered on a pixel. It is not recommended that particles larger than 10 pixels in radius (21 in diameter) be utilized in simulations so that a ratio of about 5 between system size (100^3) and individual particle size may be maintained. (If memory is available, the system size can be increased to 200^3 and larger particles used.) Using Table I, if the

Table I

Volume Occupied by Spheres of Various Radii		
Radius (pixels)	Diameter	Pixels per Sphere
1	3	19
2	9	81
3	7	179
4	9	389
5	17	739
6	13	1189
7	15	1791
8	17	2553
9	15	3695
10	21	4945

user knows the total number of pixels of a given size sphere needed, they may calculate how many spheres will be required. Since only integer numbers of spheres may be placed, some adjustment (rounding) may be required.

For this example, the aggregate occupies 20,000 pixels (2*100*100) so that 980,000 are left for the cement paste. Based on a 20% wt replacement of cement with mineral admixture, a w/s ratio of 0.45, and specific gravities of 3.2 for cement and 2.2 for the mineral admixture (silica fume), one can calculate based on equation 2 that 309,769 pixels of cement and 112,645 pixels of mineral admixture are needed. Since half of the volume of cement should be composed of each particle size (11 and 21 pixels), we should place 31 of the 21 pixel diameter particles and 212 of the 11 pixel diameter particles for the cement, for a total of 309,963 cement pixels. For the large mineral admixture particles, these numbers are 11 and 79 respectively, for a total of 112,776 mineral admixture pixels. This will result in a w/s ratio of 0.4494, very close to our desired value of 0.45. When small one-pixel mineral admixtures are to be used, we will simply add 112,776 of them to the system (to have exactly the same number of total admixture pixels as in the case of the larger particles). In order to assure that both systems hydrate to the same degree of hydration, we will specify hydration to a specified degree of hydration (70%) as opposed to hydration for some fixed number of cycles.

The data files (examples of hydrate.inp) used to execute the two simulations are as follows:

Example 1: Simulation with large mineral admixture particles

1639	random number seed
1	menu selection to add aggregate
2	aggregate thickness
2	menu selection to add spherical particles
4	number of different classes of particles to add
31	number of particles of class #1
10	radius of particles of class #1
1	phase ID of particles of class #1 (C_3S)
11	number of particles of class #2
10	radius of particles of class #2
10	phase ID of particles of class #2 (min. admixture)
212	number of particles of class #3
5	radius of particles of class #3
1	phase ID of particles of class #3 (C_3S)
79	number of particles of class #4
5	radius of particles of class #4
10	phase ID of particles of class #4 (min. admixture)
5	menu selection to measure global phase fractions
6	menu selection to measure phase fractions vs. distance from aggregate surface
4	menu selection to execute hydration
1	selection to specify desired degree of hydration
0.7	degree of hydration desired
10000	max. # of diffusion steps per cycle
0.001	maximum probability for CH nucleation
200000.	exponential scale factor for CH nucleation
5	menu selection to measure global phase fractions
6	menu selection to measure phase fractions vs. distance from aggregate surface
9	menu selection to exit program

Example 2: Simulation with one pixel mineral admixture particles

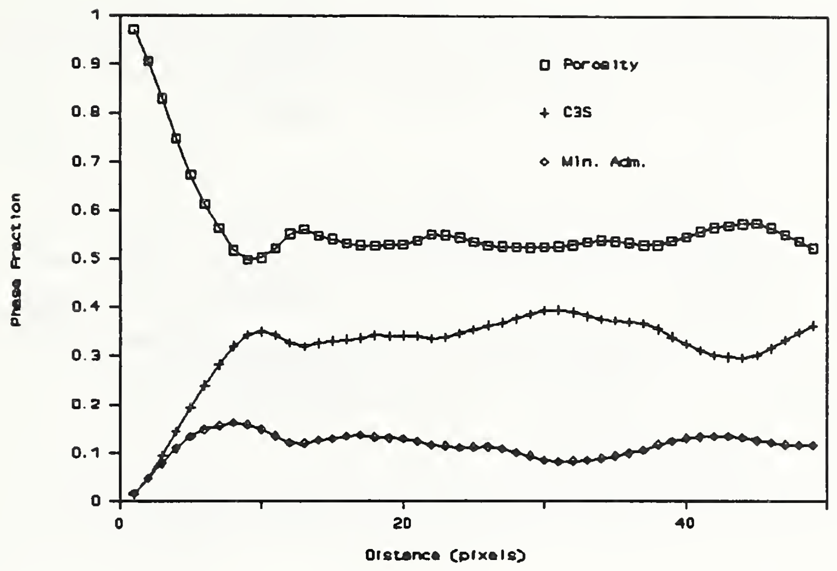
1639	random number seed
1	menu selection to add aggregate
2	aggregate thickness
2	menu selection to add spherical particles
2	number of different classes of particles to add
31	number of particles of class #1

10	radius of particles of class #1
1	phase ID of particles of class #1 (C_3S)
212	number of particles of class #2
5	radius of particles of class #2
1	phase ID of particles of class #2 (C_3S)
3	menu selection to add one-pixel particles
112776	number of one pixel particles to add
10	phase ID of one pixel particles
5	menu selection to measure global phase fractions
6	menu selection to measure phase fractions vs. distance from aggregate surface
4	menu selection to execute hydration
1	selection to specify desired degree of hydration
0.7	degree of hydration desired
10000	max. # of diffusion steps per cycle
0.001	maximum probability of CH nucleation
200000.	exponential scale factor for CH nucleation
5	menu selection to measure global phase fractions
6	menu selection to measure phase fractions vs. distance from aggregate surface
9	menu selection to exit program

The major results of this example are the phase fractions as a function of distance from the aggregate surface for the two systems. The global phase fraction data can be used to assure that 70% hydration has been achieved. The phase fraction vs. distance data were imported into a spreadsheet program to produce graphs of the various phase fractions as a function of distance for systems containing large and small mineral admixtures. These graphs are given in Figures 3, 4, and 5.

Figure 3 shows the original particle packings. As shown previously [7], the cement particles are not able to pack efficiently near the aggregate surface so that the porosity in the interfacial zone increases relative to that in the bulk paste. This also holds true for the larger mineral admixture particles. However, the smaller (one pixel) mineral admixture particles are able to pack much more efficiently than the cement particles against the aggregate surface and, since this is the highest porosity region, there are actually more mineral admixture particles in the interfacial zone than in the bulk paste for this system.

The differences in original packings for the two systems manifest themselves in different microstructures developing as hydration occurs. For systems with no mineral admixtures, it has been shown that the interfacial zone is higher in porosity and CH and lower in C-S-H and C_3S than the bulk paste [7]. The effects of mineral admixtures on this baseline microstructure are dependent on mineral admixture particle size as shown in Figure 4. Both particle sizes decrease the CH formed due to the a)



b)

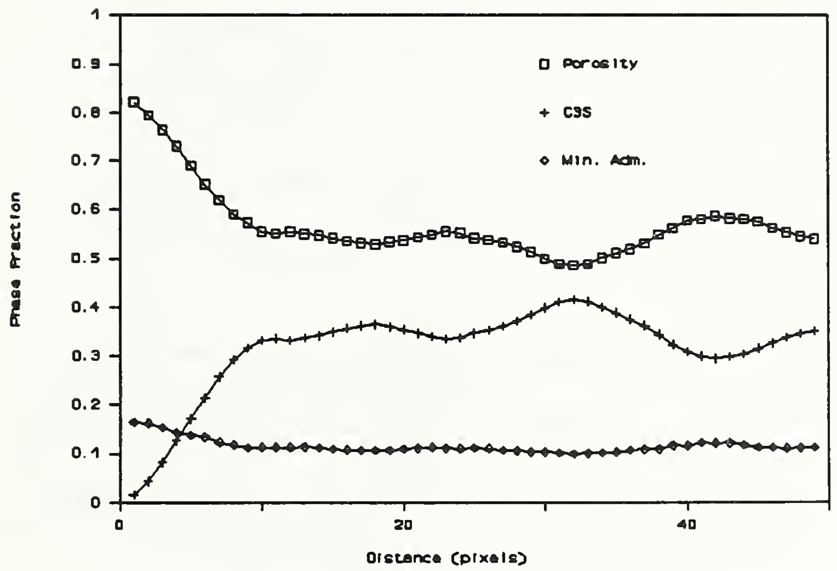
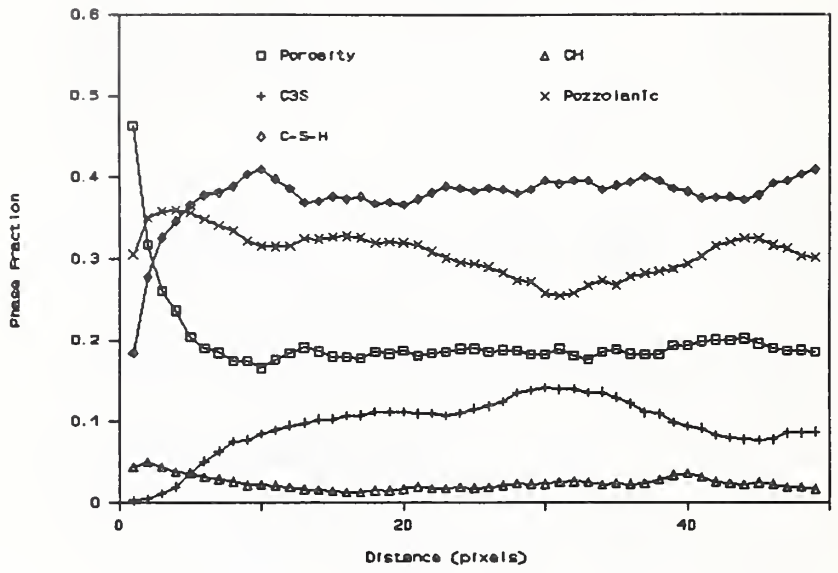


Figure 3. Original Particle Packings for a) Large and b) Small Mineral Admixture Particles

a)



b)

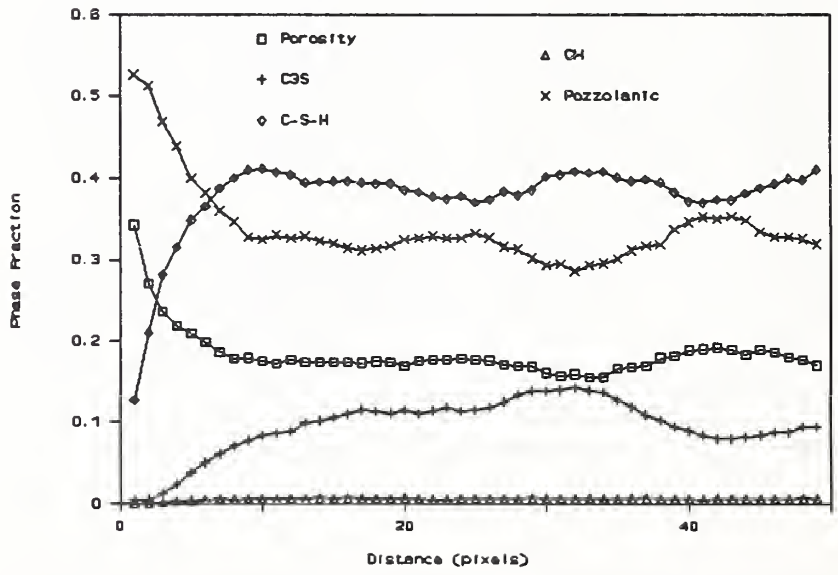


Figure 4. Phase Fractions vs. Distance from Aggregate Surface After Hydration for a) Large and b) Small Mineral Admixture Particles

occurrence of the pozzolanic reaction but a greater reduction is achieved with the smaller particles as the pozzolanic surfaces are distributed more uniformly throughout the microstructure. In fact, because with the one pixel admixture particles there is actually more pozzolanic material near the aggregate surface (Figure 3), there is also more pozzolanic C-S-H formed in the interfacial zone than in the bulk. This will aid in offsetting the deficiency in primary C-S-H (that formed directly from C_3S hydration) normally found in this zone.

To quantitatively examine this effect, the total of the C_3S, C-S-H, and pozzolanic (admixture and C-S-H) phases are plotted as a function of distance from the aggregate surface in Figure 5. From the figure, it is evident that the smaller admixture particles result in a much more homogeneous distribution of these phases as the decrease in this total phase fraction present as the aggregate surface is approached is suppressed. While the effects of admixture particle size and reactivity have been investigated in more detail [8], this simple study would suggest that adequate dispersion of silica fume is needed to maximize its performance in concrete, as the microstructure of an agglomerated system has been shown to be inferior to that of a well-dispersed one.

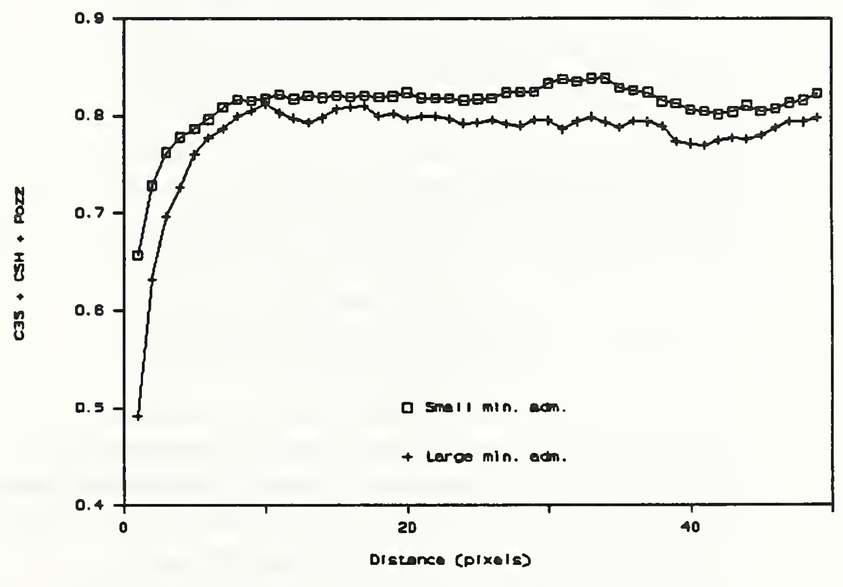


Figure 5. Total C_3S + C-S-H + Admixture Phase Fraction vs. Distance from Aggregate Surface After Hydration

4.2: EFFECT OF W/C RATIO ON CONNECTIVITY OF CAPILLARY POROSITY

For this example, we will vary the w/c ratio of ordinary cement paste and monitor the percolation or connectivity of the capillary porosity as hydration occurs. That is, we will hydrate a cement microstructure for a few cycles, measure the connectivity of the porosity phase, hydrate a few more cycles, etc. The w/c ratios employed are 0.35 and 0.5. The cement particles will consists of equal numbers of four sizes of particles, with diameters of 15, 11, 7, and 3 pixels respectively. Since each set of particles (one each of each of the four sizes) occupies 2728 pixels, 173 of each will provide a w/c ratio of 0.3497 while 141 of each will yield a w/c ratio of 0.4999. Because the hydration rate is faster during the early cycles of the hydration process, the number of cycles executed to obtain each data point will be increased as the hydration proceeds.

The data files (hydrate.inp) used to execute the two simulations are as follows:

Examples 1 (2): Simulations with w/c=0.3497 (w/c=0.4999)

8291	random number seed
2	menu selection to add cement particles
4	number of classes of particles to add
173 (141)	number of particles of class #1 to add
7	radius of particles of class #1
1	phase ID of particles of class #1 (C_3S)
173 (141)	number of particles of class #2 to add
5	radius of particles of class #2
1	phase ID of particles of class #2
173 (141)	number of particles of class #3 to add
3	radius of particles of class #3
1	phase ID of particles of class #3
173 (141)	number of particles of class #4 to add
1	radius of particles of class #4
1	phase ID of particles of class #4
5	menu selection to measure global phase fractions
7	menu selection to measure single phase connectivity
0	phase ID of which to assess percolation (porosity)
4	menu selection to execute hydration
0	selection to specify number of hydration cycles
1	number of hydration cycles to execute
10000	max. # of diffusion steps per cycle
0.001	maximum probability for CH nucleation
200000.	exponential scale factor for CH nucleation
5	menu selection to measure global phase fractions
7	menu selection to measure single phase connectivity

```
0                  phase ID of which to assess percolation (porosity)
4                  menu selection to execute hydration
0                  selection to specify number of hydration cycles
2                  number of hydration cycles to execute
5000               max. # of diffusion steps per cycle
0.001              maximum probability for CH nucleation
200000.            exponential scale factor for CH nucleation
5                  menu selection to measure global phase fractions
7                  menu selection to measure single phase connectivity
0                  phase ID of which to assess percolation (porosity)
4                  menu selection to execute hydration
0                  selection to specify number of hydration cycles
2                  number of hydration cycles to execute
5000               max. # of diffusion steps per cycle
0.001              maximum probability for CH nucleation
200000.            exponential scale factor for CH nucleation
5                  menu selection to measure global phase fractions
7                  menu selection to measure single phase connectivity
0                  phase ID of which to assess percolation (porosity)
4                  menu selection to execute hydration
0                  selection to specify number of hydration cycles
3                  number of hydration cycles to execute
5000               max. # of diffusion steps per cycle
0.001              maximum probability for CH nucleation
200000.            exponential scale factor for CH nucleation
5                  menu selection to measure global phase fractions
7                  menu selection to measure single phase connectivity
0                  phase ID of which to assess percolation (porosity)
4                  menu selection to execute hydration
0                  selection to specify number of hydration cycles
4                  number of hydration cycles to execute
5000               max. # of diffusion steps per cycle
0.001              maximum probability for CH nucleation
200000.            exponential scale factor for CH nucleation
5                  menu selection to measure global phase fractions
7                  menu selection to measure single phase connectivity
0                  phase ID of which to assess percolation (porosity)
4                  menu selection to execute hydration
0                  selection to specify number of hydration cycles
4                  number of hydration cycles to execute
5000               max. # of diffusion steps per cycle
0.001              maximum probability for CH nucleation
200000.            exponential scale factor for CH nucleation
5                  menu selection to measure global phase fractions
```

7	menu selection to measure single phase connectivity
0	phase ID of which to assess percolation (porosity)
4	menu selection to execute hydration
0	selection to specify number of hydration cycles
4	number of hydration cycles to execute
5000	max. # of diffusion steps per cycle
0.001	maximum probability for CH nucleation
200000.	exponential scale factor for CH nucleation
5	menu selection to measure global phase fractions
7	menu selection to measure single phase connectivity
0	phase ID of which to assess percolation (porosity)
4	menu selection to execute hydration
0	selection to specify number of hydration cycles
10	number of hydration cycles to execute
5000	max. # of diffusion steps per cycle
0.001	maximum probability for CH nucleation
200000.	exponential scale factor for CH nucleation
5	menu selection to measure global phase fractions
7	menu selection to measure single phase connectivity
0	phase ID of which to assess percolation (porosity)
4	menu selection to execute hydration
0	selection to specify number of hydration cycles
15	number of hydration cycles to execute
5000	max. # of diffusion steps per cycle
0.001	maximum probability for CH nucleation
200000.	exponential scale factor for CH nucleation
5	menu selection to measure global phase fractions
7	menu selection to measure single phase connectivity
0	phase ID of which to assess percolation (porosity)
4	menu selection to execute hydration
0	selection to specify number of hydration cycles
55	number of hydration cycles to execute
5000	max. # of diffusion steps per cycle
0.001	maximum probability for CH nucleation
200000.	exponential scale factor for CH nucleation
5	menu selection to measure global phase fractions
7	menu selection to measure single phase connectivity
0	phase ID of which to assess percolation (porosity)
4	menu selection to execute hydration
0	selection to specify number of hydration cycles
100	number of hydration cycles to execute
5000	max. # of diffusion steps per cycle
0.001	maximum probability for CH nucleation
200000.	exponential scale factor for CH nucleation

5	menu selection to measure global phase fractions
7	menu selection to measure single phase connectivity
0	phase ID of which to assess percolation (porosity)
9	menu selection to exit program

Based on the output from the menu selections to measure global phase fractions and to measure phase connectivity, the fraction of the total porosity which forms a connected path across the three-dimensional microstructure can be determined as a function of degree of hydration (amount of cement reacted). Figure 6 provides a plot of this connected fraction vs. degree of hydration for the two w/c ratios used in this example. The connected fraction varies from 1, as initially the

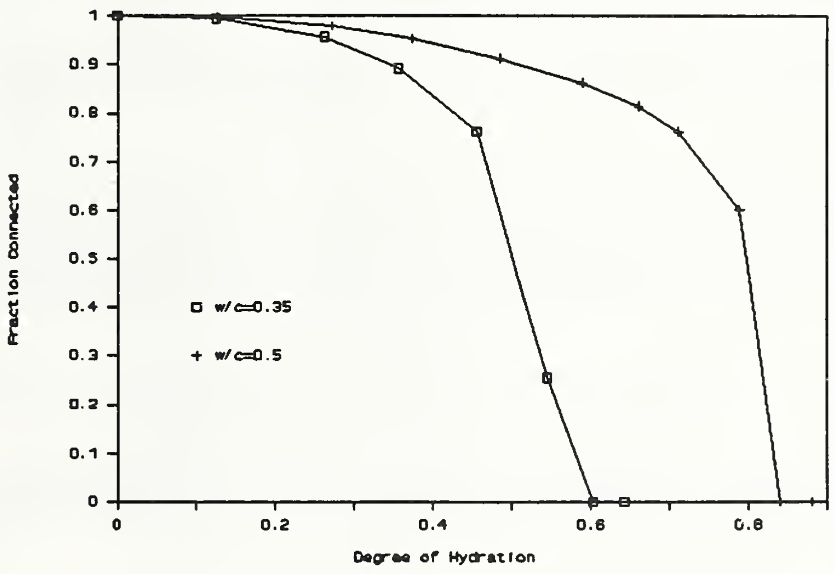


Figure 6. Fraction Connected Porosity vs. Degree of Hydration

capillary porosity is totally connected, to 0, as ultimately the hydration products fill in the porosity to an extent that discontinuity of this phase occurs. Discontinuity of the capillary porosity would be expected to have major effects on transport properties as the primary pathways for transport would shift from being the capillary pores (μm in size) to being the gel pores in the C-S-H (nm in size). Since the higher (0.5) w/c ratio contains more porosity initially, more hydration is required to achieve this discontinuity. In fact, for high enough w/c ratios (w/c $>$ 0.57), even at complete hydration, the capillary porosity remains continuous across the system [4].

Since the phase fractions of all phases are known throughout the hydration, one can also plot the fraction connected porosity vs. the total capillary porosity as shown in Figure 7. Now, the data for the two w/c ratios overlap, suggesting that it is the total capillary porosity fraction which controls the connectivity of porosity, at least for the range of w/c ratios and cement particle sizes investigated in this study.

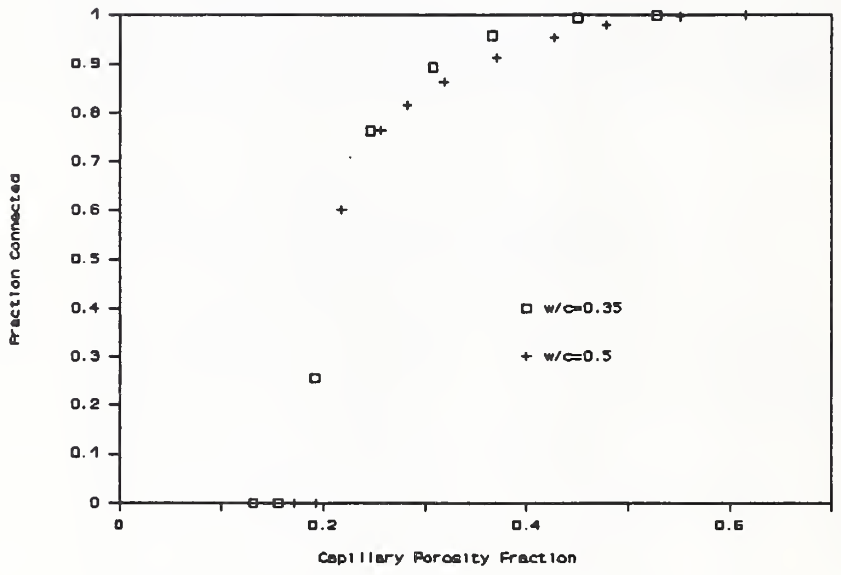


Figure 7. Fraction Connected Porosity vs. Total Porosity

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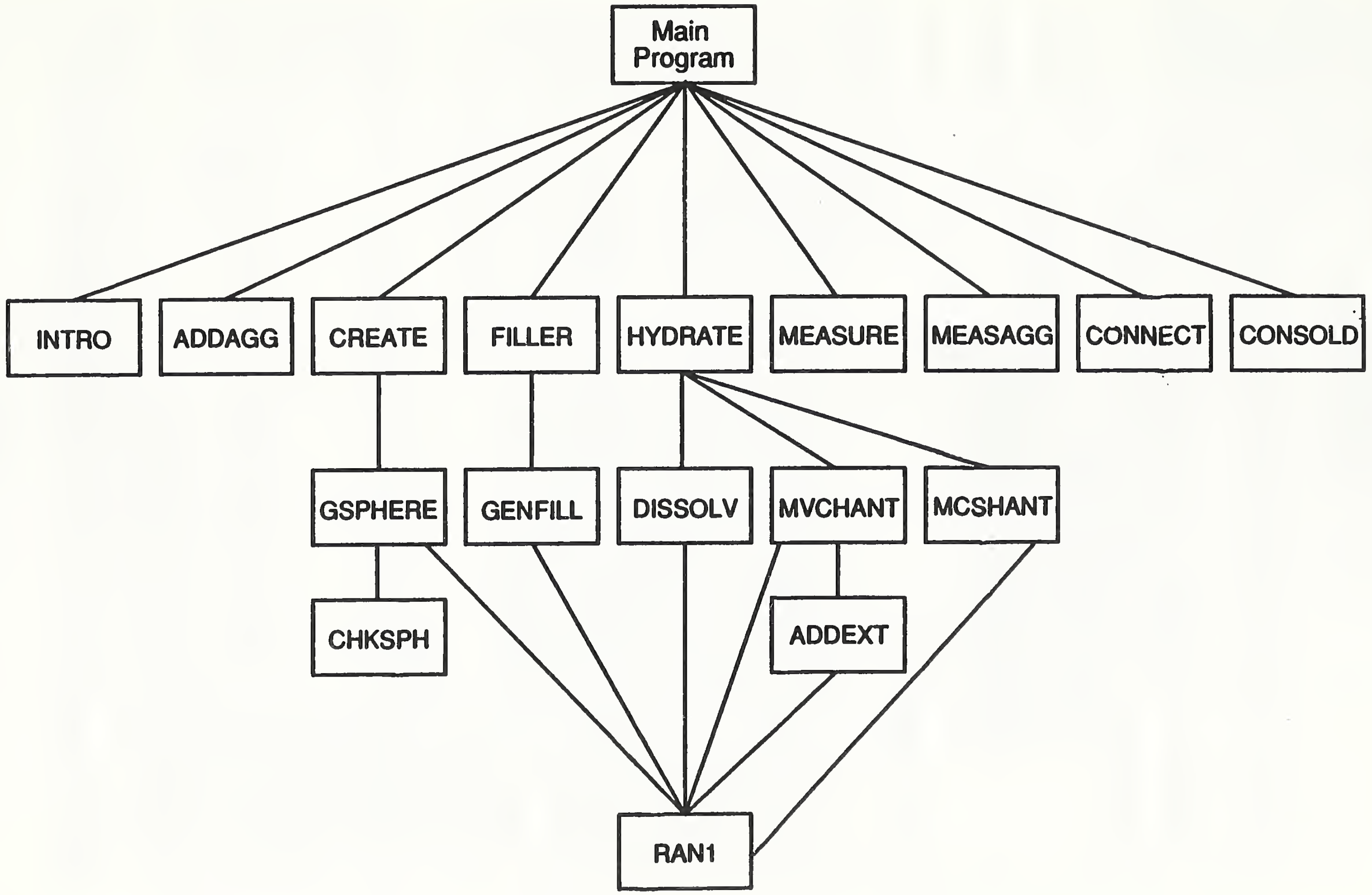
APPENDIX A
Calling Diagram for HYDRA3D



APPENDIX B
Fortran Listing for HYDRA3D

```
C   ****************************************************************
C   *                                                              *
C   * PROGRAM HYDRA3D.F : 3-D MICROSTRUCTURAL MODEL FOR CEMENT     *
C   *                                                              *
C   * DEVELOPERS: Dale P. Bentz and Edward J. Garboczi             *
C   *           Building Materials Division                        *
C   *           Building and Fire Research Laboratory              *
C   *           National Institute of Standards & Technology       *
C   *           Gaithersburg, MD  20899-0001                       *
C   *           (301)975-5865    FAX-(301)975-4032                 *
C   *                                                              *
C   * DATE: 1990-1991                                              *
C   *                                                              *
C   ****************************************************************
C    This program simulates the microstructural development of
C    cement (specifically C3S) as it hydrates.  Model is image
C    based (3-D lattice of 100*100*100 elements) and hydration is
C    modelled as a dissolution/diffusion/reaction cyclic process.
C    Tools are provided to assess phase fractions and phase
C    connectivity as the hydration proceeds.
C    Also includes the capability to add a single inert aggregate
C    to the system and assess phase fractions as a function of
C    distance from aggregate surface
C
C    PHASE ID ASSIGNMENTS
C     0- Porosity
C     1- Cement (C3S)
C     2- Diffusing Calcium hydroxide (CH) species
C     3- Diffusing Calcium silicate hydrate (CSH) species
C     4- Solid CSH
C     5- Solid CH
C     8- Aggregate
C     9- Inert filler
C     10- Pozzolanic filler/ pozzolanic CSH
C     Temporary IDs
C     6- Surface site eligible for dissolution
C     7- Burnt site in percolation routine
C
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER USERC
      INTEGER*4 ISEED
      DIMENSION R(97)
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     +AGGSIZE,NCEMENT,NCEMDIS
      COMMON /FR/IX1,IX2,IX3,R
C
C     3-D microstructure is stored in the 3-D array CEMENT
C
C
C      System size set at 100
C
      SYSSIZE=100
C
C     Clear the microstructure to all porosity
C
      DO 10 I=1,SYSSIZE
      DO 10 J=1,SYSSIZE
      DO 10 K=1,SYSSIZE
  10    CEMENT(I,J,K)=0
C
C      Call Routine to display an introduction
C
      CALL INTRO
C
C     NCEMENT is counter for cement originally present
C     NCEMDIS is counter for cement reacted so far
C     NFILL is counter of number of pozzolanic filler species
C     NPZR is counter for number of pozzolanic filler species
C       which have been consumed
C
      NCEMENT=0
      NCEMDIS=0
      NFILL=0
      NPZR=0
      AGGSIZE=0
      WRITE(6,*)'ENTER RANDOM NUMBER SEED (Integer)'
      READ(5,*)ISEED
      WRITE(6,*)ISEED
C
C     Present user with menu of choices and execute
C     appropriate option until user elects to stop
C
```

```
 40   WRITE(6,*)'INPUT USER CHOICE'
      WRITE(6,*)' 1) ADD A FLAT INERT AGGREGATE TO MICROSTRUCTURE'
      WRITE(6,*)' 2) ADD SPHERICAL PARTICLES TO MICROSTRUCTURE'
      WRITE(6,*)' 3) ADD ONE-PIXEL FILLER PARTICLES TO MICROSTRUCTURE'
      WRITE(6,*)' 4) HYDRATE MICROSTRUCTURE'
      WRITE(6,*)' 5) MEASURE PHASE FRACTIONS'
      WRITE(6,*)' 6) MEASURE PHASE FRACTIONS AS A FUNCTION'
      WRITE(6,*)'    OF DISTANCE FROM AGGREGATE SURFACE'
      WRITE(6,*)' 7) MEASURE SINGLE PHASE CONNECTIVITY'
      WRITE(6,*)' 8) MEASURE TOTAL SOLIDS CONNECTIVITY'
      WRITE(6,*)' 9) EXIT'
      READ(5,*)USERC
      WRITE(6,*)USERC
      IF(USERC.EQ.1) THEN
        CALL ADDAGG
      ELSE IF(USERC.EQ.2) THEN
        CALL CREATE
      ELSE IF (USERC.EQ.3) THEN
       CALL FILLER
      ELSE IF (USERC.EQ.4) THEN
       CALL HYDRATE
      ELSE IF (USERC.EQ.5) THEN
       CALL MEASURE
      ELSE IF (USERC.EQ.6) THEN
        CALL MEASAGG
      ELSE IF (USERC.EQ.7) THEN
       CALL CONNECT
      ELSE IF (USERC.EQ.8) THEN
       CALL CONSOLD
      ELSE IF (USERC.EQ.9) THEN
        GOTO 50
      ENDIF
       GOTO 40
   50 STOP
      END
      FUNCTION RAN1(IDUM)
C
C      Portable random number generator, RAN1
C      To generate uniform random deviates between 0 and 1.0
C      From: Numerical Recipes in Fortran
C           Press, Flannery, Teukolsky, and Vetterling
C
      DIMENSION R(97)
      PARAMETER (M1=259200,IA1=7141,IC1=54773,RM1=1./M1)
      PARAMETER (M2=134456,IA2=8121,IC2=28411,RM2=1./M2)
      PARAMETER (M3=243000,IA3=4561,IC3=51349)
      COMMON /FR/IX1,IX2,IX3,R
      DATA IFF /0/

      IF(IDUM.LT.0.OR.IFF.EQ.0) THEN
        IFF=1
        IX1=MOD(IC1-IDUM,M1)
        IX1=MOD(IA1*IX1+IC1,M1)
        IX2=MOD(IX1,M2)
        IX1=MOD(IA1*IX1+IC1,M1)
        IX3=MOD(IX1,M3)
        DO 11 J=1,97
          IX1=MOD(IA1*IX1+IC1,M1)
          IX2=MOD(IA2*IX2+IC2,M2)
          R(J)=(FLOAT(IX1)+FLOAT(IX2)*RM2)*RM1
 11     CONTINUE
        IDUM=1
      ENDIF
      IX1=MOD(IA1*IX1+IC1,M1)
      IX2=MOD(IA2*IX2+IC2,M2)
      IX3=MOD(IA3*IX3+IC3,M3)
      J=1+(97*IX3)/M3
      IF(J.GT.97.OR.J.LT.1) THEN
        WRITE(6,*)'ERROR IN RANDOM NUMBER GENERATOR'
      ENDIF
      RAN1=R(J)
      R(J)=(FLOAT(IX1)+FLOAT(IX2)*RM2)*RM1
      RETURN
      END
      SUBROUTINE INTRO
C
C      Subroutine INTRO to display an introduction to HYDRA3D
C      Called by: Main Program
C
      WRITE(6,*)'Welcome to HYDRA3D, a digital-image-based cement'
      WRITE(6,*)'microstructural model.  This program has been '
      WRITE(6,*)'developed to simulate the microstructural '
      WRITE(6,*)'development of cement, specifically tricalcium'
      WRITE(6,*)'silicate, C3S, as it reacts with water.  In '
      WRITE(6,*)'addition to C3S, the user may also add a single'
      WRITE(6,*)'inert aggregate and/or inert or pozzolanic mineral'
      WRITE(6,*)'admixture particles to the microstructure.  In'
      WRITE(6,*)'addition to hydration, the user may also assess'
      WRITE(6,*)'phase fractions (either globally or as a function'
      WRITE(6,*)'of distance from the aggregate surface) and '
      WRITE(6,*)'the connectivity of any individual phase or of'
      WRITE(6,*)'the total solids (neglecting any aggregate) present'
      WRITE(6,*)'in the system.'
      WRITE(6,*)'  '
      RETURN
      END
      SUBROUTINE ADDAGG
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     + AGGSIZE,NCEMENT,NCEMDIS
      INTEGER IX,IY,IZ
C
C      Subroutine ADDAGG to add a flat aggregate of user-specified
C      thickness to microstructure
C      Called by: Main program
C
      WRITE(6,*)'ENTER AGGREGATE THICKNESS (even integer)'
      READ(5,*)AGGSIZE
      WRITE(6,*)AGGSIZE

      DO 45 IX=((SYSSIZE-AGGSIZE+2)/2),((SYSSIZE+AGGSIZE)/2)
      DO 45 IY=1,SYSSIZE
      DO 45 IZ=1,SYSSIZE
 45      CEMENT(IX,IY,IZ)=8
      RETURN
      END
      SUBROUTINE CREATE
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     + AGGSIZE,NCEMENT,NCEMDIS
      INTEGER NUMSIZE,SPHNUM(10),SPHRAD(10),SPHID(10)
C
C       Subroutine CREATE to obtain user input to create a starting system
C       consisting of digitized spheres of various radii and phase IDs
C       Called by: Main program
C       Calls: Subroutine GSPHERE
C
 11   WRITE(6,*)'ENTER NUMBER OF DIFFERENT SIZE SPHERES TO USE(MAX 10)'
```

```
      READ(5,*)NUMSIZE
      IF(NUMSIZE.LT.1.OR.NUMSIZE.GT.10) THEN
        GOTO 11
      ENDIF
      WRITE(6,*)NUMSIZE
      WRITE(6,*)'ENTER NUMBER, SIZE, AND PHASE FOR EACH CLASS'
      WRITE(6,*)'(LARGEST RADIUS 1ST)'
      DO 21 I=1,NUMSIZE
        WRITE(6,*)'ENTER NUMBER OF SPHERES OF CLASS',I
        READ(5,*)SPHNUM(I)
         WRITE(6,*)SPHNUM(I)
        WRITE(6,*)'ENTER RADIUS OF SPHERES OF CLASS',I
        WRITE(6,*)'(Integer <=10 PLEASE)'
        READ(5,*)SPHRAD(I)
        WRITE(6,*)SPHRAD(I)
        WRITE(6,*)'ENTER PHASE ID TO BE ASSIGNED TO SPHERES OF CLASS',I
        WRITE(6,*)'(1- C3S, 9- Inert filler  10- Pozzolanic filler'
        READ(5,*)SPHID(I)
  21    WRITE(6,*)SPHID(I)
C
      CALL GSPHERE(NUMSIZE,SPHNUM,SPHRAD,SPHID)
      RETURN
      END
      SUBROUTINE GSPHERE(NUMGEN,NUMEACH,SIZEEACH,PHEACH)
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     +AGGSIZE,NCEMENT,NCEMDIS
      INTEGER NUMGEN,NUMEACH(10),SIZEEACH(10),PHEACH(10),RADIUS
      INTEGER PHSPH,COUNT,X,Y,Z
      REAL RX,RY,RZ
C
C      Subroutine GSPHERE to generate spheres of a fixed number of size
C      classes
C      Input: Number of size classes, number, size, and ID of spheres
C             in each size class
C      Called by: Subroutine CREATE
C      Calls: Subroutine CHKSPH
C
C      Generate the requested number of each size of sphere
C
      DO 131 I=1,NUMGEN
      RADIUS=SIZEEACH(I)
      PHSPH=PHEACH(I)
      DO 131 J=1,NUMEACH(I)
 20    COUNT=0
C
C     Generate a random center location for this sphere
C
       RX=RAN1(ISEED)
       RY=RAN1(ISEED)
       RZ=RAN1(ISEED)
       X=SYSSIZE*RX+1
       Y=SYSSIZE*RY+1
       Z=SYSSIZE*RZ+1
C
C      Call routine to check is sphere can be placed at chosen location
C
       CALL CHKSPH(COUNT,X,Y,Z,RADIUS,1,PHSPH)
      IF(COUNT.NE.0) THEN
       GOTO 20
      ENDIF
C
C     Place the sphere at the selected location
C
 131    CALL CHKSPH(COUNT,X,Y,Z,RADIUS,2,PHSPH)
      RETURN
      END
      SUBROUTINE CHKSPH(SFLG,XIN,YIN,ZIN,RADD,WFLG,PHID)
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     +AGGSIZE,NCEMENT,NCEMDIS
      INTEGER SFLG,XIN,YIN,ZIN,RADD,WFLG,PHID,NOFITS
      INTEGER XP,YP,ZP
      REAL DIST
C
C     Subroutine CHKSPH to either check or perform sphere location
C     Inputs: Flag used to send back indication if sphere fits,
C            x,y, and z coordinates of sphere center, radius of
C            sphere, and flag indicating if sphere is to be
C            checked (1) or placed (2)
C     Called by: Subroutine GSPHERE
C
      NOFITS=0
C
```

```
C     Check all pixels within the sphere
C
      DO 221 I=XIN-RADD,XIN+RADD
      DO 221 J=YIN-RADD,YIN+RADD
      DO 221 K=ZIN-RADD,ZIN+RADD
      XP=I
      YP=J
      ZP=K
C
C     Use periodic boundaries to wrap a sphere from one side of
C     the 3-D system to the other
C
      IF(XP.LT.1) THEN
        XP=SYSSIZE+XP
      ENDIF
      IF(XP.GT.SYSSIZE) THEN
        XP=XP-SYSSIZE
      ENDIF
      IF(YP.LT.1) THEN
        YP=SYSSIZE+YP
      ENDIF
      IF(YP.GT.SYSSIZE) THEN
        YP=YP-SYSSIZE
      ENDIF
      IF(ZP.LT.1) THEN
        ZP=SYSSIZE+ZP
      ENDIF
      IF(ZP.GT.SYSSIZE) THEN
        ZP=ZP-SYSSIZE
      ENDIF
C
C     Compute the distance from the center of the sphere to this point
C
      DIST=SQRT(FLOAT((I-XIN)*(I-XIN)+(J-YIN)*(J-YIN)+
     &(K-ZIN)*(K-ZIN)))
      IF((DIST-0.5).LE.RADD) THEN
        IF(WFLG.EQ.2) THEN
          CEMENT(XP,YP,ZP)=PHID
C
C     Update counter of C3S species if necessary
C
          IF(PHID.EQ.1) THEN
            NCEMENT=NCEMENT+1
          ENDIF
```

```
C
C     Update counter of pozzolanic filler species if necessary
C
          IF(PHID.EQ.10) THEN
            NFILL=NFILL+1
          ENDIF
        ENDIF
        IF(WFLG.EQ.1.AND.CEMENT(XP,YP,ZP).NE.0) THEN
C
C       Sphere can not be fit at present location without overlapping
C       some other sphere so notify calling routine of lack of fit
C
          NOFITS=1
          GOTO 231
        ENDIF
      ENDIF
 221    CONTINUE
 231   SFLG=NOFITS
      RETURN
      END
      SUBROUTINE FILLER
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     +AGGSIZE,NCEMENT,NCEMDIS
      INTEGER FILLID,NADD
C
C     Subroutine FILLER to obtain user input as to number and type of
C     filler particles
C     Called by: Main program
C     Calls: Subroutine GENFILL
C
      WRITE(6,*)'ENTER NUMBER OF FILLER PARTICLES TO ADD'
      READ(5,*)NADD
      WRITE(6,*)NADD
      WRITE(6,*)'ENTER FILLER ID TO USE (10-POZZOLANIC, 9-INERT)'
      READ(5,*)FILLID
      WRITE(6,*)FILLID
      IF(FILLID.NE.9.AND.FILLID.NE.10) THEN
        STOP
      ENDIF
      CALL GENFILL(NADD,FILLID)
      IF(FILLID.EQ.10) THEN
```

```
        NFILL=NFILL+NADD
      ENDIF
      RETURN
      END
      SUBROUTINE GENFILL(NTOPL,FILVAL)
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     +AGGSIZE,NCEMENT,NCEMDIS
      INTEGER NTOPL,FILVAL,PFILL,XFILL,YFILL,ZFILL
      REAL RXF,RYF,RZF
C
C      Subroutine GENFILL to locate one pixel filler particles at random
C      unoccupied locations in the 3-D microstructure
C      Input: Number and ID of filler particles to generate
C      Called by: Subroutine FILLER
C
      DO 683 I=1,NTOPL
      PFILL=0
C
C     PFILL indicates successful placement of this filler particle
C
C     Generate a random location and attempt to place filler particle
C     there
C
 684  RXF=RAN1(ISEED)
      RYF=RAN1(ISEED)
      RZF=RAN1(ISEED)
      XFILL=SYSSIZE*RXF+1
      YFILL=SYSSIZE*RYF+1
      ZFILL=SYSSIZE*RZF+1
C
      IF(CEMENT(XFILL,YFILL,ZFILL).EQ.0) THEN
        PFILL=1
        CEMENT(XFILL,YFILL,ZFILL)=FILVAL
      ENDIF
      IF(PFILL.EQ.0) THEN
        GOTO 684
      ENDIF
 683  CONTINUE
      RETURN
      END
      SUBROUTINE HYDRATE
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     + AGGSIZE,NCEMENT,NCEMDIS
      INTEGER NITER,NUMANTS,ANTSX(500000),ANTSY(500000)
      INTEGER ANTSZ(500000),XANT,YANT,ZANT,NUMLEFT,NBLEFT,ALIVE
      INTEGER NBLUE,ANTTYPE,STOPCH
      REAL NUCPROB,NUCSCALE,BETERM,BBIAS,ALPHA,ALMAX
C
C      Subroutine HYDRATE to obtain user input and control execution
C      of a number of hydration cycles
C      Called by: MAIN program
C      Calls: Subroutines DISSOLV, MVCHANT, MCSHANT
C
C      NITER is number of hydration cycles to perform
C      NTIMES is number of random steps diffused per cycle
C            before all diffusing species convert to solid
C            at current location
C      NUCPROB and NUCSCALE are parameters to control the number
C             and size of nucleating CH crystals by controlling
C             the nucleation probability
C
      WRITE(6,*)'DO YOU WISH TO SPECIFY 0) MAX. # OF CYCLES OR'
      WRITE(6,*)'1) MAXIMUM DEGREE OF HYDRATION'
      READ(5,*)STOPCH
      IF(STOPCH.EQ.0) THEN
        WRITE(6,*)'ENTER NO. OF CYCLES (ITERATIONS) TO PERFORM'
        READ(5,*)NITER
        WRITE(6,*)NITER
        ALMAX=1.0
      ENDIF
      IF(STOPCH.EQ.1) THEN
        WRITE(6,*)'ENTER DESIRED DEGREE OF HYDRATION'
        READ(5,*)ALMAX
        WRITE(6,*)ALMAX
        NITER=5000
      ENDIF
      WRITE(6,*)'ENTER MAX. NUMBER OF DIFFUSION STEPS PER CYCLE'
      READ(5,*)NTIMES
      WRITE(6,*)NTIMES
      WRITE(6,*)'ENTER MAXIMUM PROBABILITY FOR CH NUCLEATION (0.0-1.0)'
      READ(5,*)NUCPROB
      WRITE(6,*)NUCPROB
      WRITE(6,*)'ENTER EXPONENTIAL SCALE FACTOR FOR CH NUCLEATION'
      READ(5,*)NUCSCALE
      WRITE(6,*)NUCSCALE
C
C      Loop for each hydration cycle
C
      ALPHA=FLOAT(NCEMDIS)/FLOAT(NCEMENT)
      DO 22 I=1,NITER
      IF(ALPHA.GE.ALMAX) THEN
        GOTO 28
      ENDIF
C
C      NUMANTS is counter for number of diffusing species generated
C
          NUMANTS=0
        CALL DISSOLV(ANTSX,ANTSY,ANTSZ,NUMANTS)
        NUMLEFT=NUMANTS
        ALPHA=FLOAT(NCEMDIS)/FLOAT(NCEMENT)
        WRITE(6,*)'NUMBER DISSOLVED=  ',NUMANTS
C
C       NBLUE is counter for number of diffusing CH species
C
        NBLUE=61*NUMANTS/231
C
C        Loop for each diffusion step
C
        DO 32 J=1,NTIMES
        CYCLENO=J
C
C        If no diffusing species remain then go to next dissolution
C
        IF(NUMLEFT.EQ.0) THEN
          GOTO 22
        ENDIF
        NUMLEFT=0
        NBLEFT=0
C
C        Compute probability of CH nucleation based on current system
C
        BETERM=EXP(-FLOAT(NBLUE)/NUCSCALE)
        BBIAS=NUCPROB*(1.-BETERM)
C
C        Loop for each diffusing species remaining
C
```

```
       DO 52 K=1,NUMANTS
C
C      Obtain the location and type of this diffusing species
C
        XANT=ANTSX(K)
        YANT=ANTSY(K)
        ZANT=ANTSZ(K)
        ANTTYPE=CEMENT(XANT,YANT,ZANT)
C
C       ALIVE indicates if species is still diffusing or has
C            become solid
C
        ALIVE=1
         IF((ANTTYPE.NE.2).AND.(ANTTYPE.NE.3)) THEN
           WRITE(6,*)'Error- Cement pixel is ',ANTTYPE
        ENDIF
        IF(ANTTYPE.EQ.2) THEN
C        Call routine to move a CH diffusing species
          CALL MVCHANT(XANT,YANT,ZANT,ALIVE,BBIAS)
        ENDIF
         IF(ANTTYPE.EQ.3) THEN
C        Call routine to move a CSH diffusing species
          CALL MCSHANT(XANT,YANT,ZANT,ALIVE)
        ENDIF
        IF(ALIVE.EQ.1) THEN
          NUMLEFT=NUMLEFT+1
C
C        Store new location of diffusing species if still alive
C
          ANTSX(NUMLEFT)=XANT
          ANTSY(NUMLEFT)=YANT
          ANTSZ(NUMLEFT)=ZANT
         IF(ANTTYPE.EQ.2) THEN
           NBLEFT=NBLEFT+1
          ENDIF
        ENDIF
 52     CONTINUE
      NUMANTS=NUMLEFT
      NBLUE=NBLEFT
 32   CONTINUE
 22   CONTINUE
 28   RETURN
     END
     SUBROUTINE DISSOLV(DISX,DISY,DISZ,NUMDIS)
```

```
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     +AGGSIZE,NCEMENT,NCEMDIS
      INTEGER DISX(500000),DISY(500000),DISZ(500000),NUMDIS
      INTEGER XC,YC,ZC,NEDGE,NCHADD,NCSHADD,NUMORG
      REAL RDIS,RDX,RDY,RDZ
      INTEGER IRND
C
C      Subroutine DISSOLV to perform dissolution at beginning of hydration
C      cycle
C      Returns arrays of x,y, and z coordinates of generated diffusing
C      species and number of species generated
C      Called by: Subroutine HYDRATE
C
      NUMDIS=0
C
C       HIGHLIGHT ALL C3S EDGE POINTS
C
      DO 15 I=1,SYSSIZE
      DO 15 J=1,SYSSIZE
      DO 15 K=1,SYSSIZE
      IF(CEMENT(I,J,K).EQ.1) THEN
        NEDGE=1
C
C      Check all six neighbors in 3-D to see if pixel is on edge
C      Note that periodic boundaries are employed throughout
C
        XC=I-1
        IF(XC.LT.1) XC=SYSSIZE
        NEDGE=NEDGE*CEMENT(XC,J,K)
        XC=I+1
        IF(XC.GT.SYSSIZE) XC=1
        NEDGE=NEDGE*CEMENT(XC,J,K)
        YC=J-1
        IF(YC.LT.1) YC=SYSSIZE
        NEDGE=NEDGE*CEMENT(I,YC,K)
        YC=J+1
        IF(YC.GT.SYSSIZE) YC=1
        NEDGE=NEDGE*CEMENT(I,YC,K)
        ZC=K-1
        IF(ZC.LT.1) ZC=SYSSIZE
        NEDGE=NEDGE*CEMENT(I,J,ZC)
        ZC=K+1
        IF(ZC.GT.SYSSIZE) ZC=1
        NEDGE=NEDGE*CEMENT(I,J,ZC)
C
C        If on edge, assign temporary ID for second stage
C
        IF(NEDGE.EQ.0) CEMENT(I,J,K)=6
      ENDIF
 15   CONTINUE
C
C       RANDOMLY DISSOLVE ALL EDGE POINTS
C
      DO 25 I=1,SYSSIZE
      DO 25 J=1,SYSSIZE
      DO 25 K=1,SYSSIZE
      IF(CEMENT(I,J,K).EQ.6) THEN
C
C      Dissolution is performed by having pixel of interest
C      execute a one-step random walk and seeing if it steps
C      into pore space
C
        RDIS=RAN1(ISEED)
        IRND=6*RDIS+1
        XC=I
        YC=J
        ZC=K
        IF(IRND.EQ.1) THEN
          XC=XC-1
          IF(XC.LT.1) XC=SYSSIZE
        ENDIF
        IF(IRND.EQ.2) THEN
          XC=XC+1
          IF(XC.GT.SYSSIZE) XC=1
        ENDIF
        IF(IRND.EQ.3) THEN
          YC=YC-1
          IF(YC.LT.1) YC=SYSSIZE
        ENDIF
        IF(IRND.EQ.4) THEN
          YC=YC+1
          IF(YC.GT.SYSSIZE) YC=1
        ENDIF
        IF(IRND.EQ.5) THEN
          ZC=ZC-1
        IF(ZC.LT.1) ZC=SYSSIZE
       ENDIF
       IF(IRND.EQ.6) THEN
        ZC=ZC+1
        IF(ZC.GT.SYSSIZE) ZC=1
       ENDIF
       IF(CEMENT(XC,YC,ZC).EQ.0) THEN
C
C       Generate a diffusing C-S-H species at the new location
C
        CEMENT(XC,YC,ZC)=3
        CEMENT(I,J,K)=0
        NUMDIS=NUMDIS+1
        DISX(NUMDIS)=XC
        DISY(NUMDIS)=YC
        DISZ(NUMDIS)=ZC
       ELSE
        CEMENT(I,J,K)=1
       ENDIF
      ENDIF
 25  CONTINUE
C
C      ADD IN EXTRA DIFFUSING SPECIES
C
      WRITE(6,*)'ORIGINAL DISSOLVED=  ',NUMDIS
      NUMORG=NUMDIS
      NCEMDIS=NCEMDIS+NUMDIS
C
C     Expansion factors 0.7 for C-S-H and 0.61 for CH are
C     taken from data of Young & Hansen
C      MRS Proceedings, Vol. 85, pp. 313-22, 1987.
C
      NCSHADD=0.7*FLOAT(NUMDIS)
      NCHADD=0.61*FLOAT(NUMDIS)
      NUMDIS=NUMDIS+NCSHADD+NCHADD
      IF(NUMDIS.GT.500000) THEN
        WRITE(6,*)'Too many dissolved species created'
        WRITE(6,*)'Aborting execution'
        STOP
      ENDIF
      DO 35 I=1,NCHADD+NCSHADD
C
C     Locate extra diffusing species at random unoccupied
C     sites in the pore space
```

```
C
 45     RDX=RAN1(ISEED)
       RDY=RAN1(ISEED)
       RDZ=RAN1(ISEED)
       XC=SYSSIZE*RDX+1
       YC=SYSSIZE*RDY+1
       ZC=SYSSIZE*RDZ+1
       IF(CEMENT(XC,YC,ZC).EQ.0) THEN
         DISX(NUMORG+I)=XC
         DISY(NUMORG+I)=YC
         DISZ(NUMORG+I)=ZC
         IF(I.LE.NCHADD) THEN
           CEMENT(XC,YC,ZC)=2
         ELSE
           CEMENT(XC,YC,ZC)=3
         ENDIF
       ELSE
         GOTO 45
      ENDIF
 35    CONTINUE
      RETURN
      END
      SUBROUTINE MVCHANT(XMCH,YMCH,ZMCH,CHYET,PNUC)
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     +AGGSIZE,NCEMENT,NCEMDIS
      INTEGER XMCH,YMCH,ZMCH,CHYET,XM,YM,ZM,MGEN
      REAL PNUC,PGEN
      REAL RM
C
C     Subroutine MVCHANT to move a diffusing CH species and check
C      for nucleation or surface reaction
C     Inputs: Current location of diffusing species and
C             current probability for nucleation (PNUC)
C     Returns: Flag indicating if reaction has occurred (CHYET)
C     Called by: Subroutine HYDRATE
C     Calls: Subroutine ADDEXT
C
C
C      FIRST CHECK FOR NUCLEATION
C      If last diffusion step in this cycle, convert to solid CH
C
```

```
      PGEN=RAN1(ISEED)
      IF((PNUC.GT.PGEN).OR.(CYCLENO.EQ.NTIMES)) THEN
        CEMENT(XMCH,YMCH,ZMCH)=5
        CHYET=0
      ELSE
C       GENERATE A RANDOM MOVE
        RM=RAN1(ISEED)
        MGEN=6*RM+1
        XM=XMCH
        YM=YMCH
        ZM=ZMCH
        IF(MGEN.EQ.1) THEN
          XM=XM-1
          IF(XM.LT.1) XM=SYSSIZE
        ENDIF
        IF(MGEN.EQ.2) THEN
          XM=XM+1
          IF(XM.GT.SYSSIZE) XM=1
        ENDIF
        IF(MGEN.EQ.3) THEN
          YM=YM-1
          IF(YM.LT.1) YM=SYSSIZE
        ENDIF
        IF(MGEN.EQ.4) THEN
          YM=YM+1
          IF(YM.GT.SYSSIZE) YM=1
        ENDIF
        IF(MGEN.EQ.5) THEN
          ZM=ZM-1
          IF(ZM.LT.1) ZM=SYSSIZE
        ENDIF
        IF(MGEN.EQ.6) THEN
          ZM=ZM+1
          IF(ZM.GT.SYSSIZE) ZM=1
        ENDIF
C
C       Check for surface reaction
C
        IF(CEMENT(XM,YM,ZM).EQ.5) THEN
          CEMENT(XMCH,YMCH,ZMCH)=5
          CHYET=0
        ENDIF
C
C       Check for pozzolanic reaction of diffusing CH
```

```
C     Each pozzolanic filler particle may react with
C      up to 2.08 CH diffusing species
C     Assumes pozzolanic filler is pure silica with a
C     molar volume of 27 cm^3/mole
C
      XNFILL=NFILL
      IF((CEMENT(XM,YM,ZM).EQ.10).AND.(NPZR.LE.(INT(2.08*XNFILL))))
     & THEN
        CEMENT(XMCH,YMCH,ZMCH)=10
        NPZR=NPZR+1
        CHYET=0
        RM=RAN1(ISEED)
C
C      In pozzolanic reaction there is a probability of 0.73 that
C       two pixels of product should be produced instead of one
C
        IF(RM.LE.(0.73)) THEN
        CALL ADDEXT(XMCH,YMCH,ZMCH)
        ENDIF
      ENDIF
C
C     If new location is pore space, perform the move
C
      IF(CEMENT(XM,YM,ZM).EQ.0) THEN
        CEMENT(XM,YM,ZM)=2
        CEMENT(XMCH,YMCH,ZMCH)=0
        XMCH=XM
        YMCH=YM
        ZMCH=ZM
      ENDIF
    ENDIF
    RETURN
    END
    SUBROUTINE ADDEXT(XINN,YINN,ZINN)
    INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
    INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
    INTEGER*4 ISEED
    COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
   +AGGSIZE,NCEMENT,NCEMDIS
    INTEGER XINN,YINN,ZINN,XCHR,YCHR,ZCHR,FCHR,MGEN1
    INTEGER TFACT,TRIED(6)
    REAL RMADD
C
C    Subroutine ADDEXT to add in extra pozzolanic CSH when CH
```

```
C     reacts with silica fume (pozzolanic filler)
C     Inputs: x,y, and z coordinates of new pozz. CSH
C     Called by: Subroutine MVCHANT
C
C     ADD IN EXTRA POZZOLANIC CSH TO ACCOUNT FOR VOLUME BALANCE
C     RANDOM TRIES AT NEIGHBORING LOCATIONS (6) UNTIL SUCCESSFUL
C     OR ALL 6 HAVE BEEN TRIED
C
      DO 93 I1=1,6
 93   TRIED(I1)=0
      FCHR=0
      TFACT=0
 88   RMADD=RAN1(ISEED)
      MGEN1=6*RMADD+1
      XCHR=XINN
      YCHR=YINN
      ZCHR=ZINN
      IF((MGEN1.EQ.1).AND.(TRIED(1).EQ.0)) THEN
        XCHR=XCHR-1
         TRIED(1)=1
         TFACT=TFACT+1
        IF(XCHR.LT.1) XCHR=SYSSIZE
      ENDIF
      IF((MGEN1.EQ.2).AND.(TRIED(2).EQ.0)) THEN
        XCHR=XCHR+1
         TRIED(2)=1
         TFACT=TFACT+1
        IF(XCHR.GT.SYSSIZE) XCHR=1
      ENDIF
      IF((MGEN1.EQ.3).AND.(TRIED(3).EQ.0)) THEN
        YCHR=YCHR-1
         TRIED(3)=1
         TFACT=TFACT+1
        IF(YCHR.LT.1) YCHR=SYSSIZE
      ENDIF
      IF((MGEN1.EQ.4).AND.(TRIED(4).EQ.0)) THEN
        YCHR=YCHR+1
         TRIED(4)=1
         TFACT=TFACT+1
        IF(YCHR.GT.SYSSIZE) YCHR=1
      ENDIF
      IF((MGEN1.EQ.5).AND.(TRIED(5).EQ.0)) THEN
        ZCHR=ZCHR-1
         TRIED(5)=1
        TFACT=TFACT+1
       IF(ZCHR.LT.1) ZCHR=SYSSIZE
      ENDIF
       IF((MGEN1.EQ.6).AND.(TRIED(6).EQ.0)) THEN
       ZCHR=ZCHR+1
        TRIED(6)=1
        TFACT=TFACT+1
       IF(ZCHR.GT.SYSSIZE) ZCHR=1
      ENDIF
      IF(CEMENT(XCHR,YCHR,ZCHR).EQ.0) THEN
        CEMENT(XCHR,YCHR,ZCHR)=10
         FCHR=1
        GOTO 89
      ENDIF
      IF(TFACT.EQ.6) GOTO 89
      GOTO 88
  89  IF(FCHR.EQ.0) THEN
C
C       TRY EXTRA CSH AT RANDOM LOCATIONS IN PORE SPACE
C       UNTIL IT FINDS AN EMPTY SITE
C
      RMADD=RAN1(ISEED)
      XCHR=SYSSIZE*RMADD+1
      RMADD=RAN1(ISEED)
      YCHR=SYSSIZE*RMADD+1
      RMADD=RAN1(ISEED)
      ZCHR=SYSSIZE*RMADD+1
      IF(CEMENT(XCHR,YCHR,ZCHR).EQ.0) THEN
        CEMENT(XCHR,YCHR,ZCHR)=10
        FCHR=1
      ENDIF
      GOTO 89
      ENDIF
      RETURN
      END
      SUBROUTINE MCSHANT(XMCSH,YMCSH,ZMCSH,CSHYET)
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     +AGGSIZE,NCEMENT,NCEMDIS
      INTEGER XMCSH,YMCSH,ZMCSH,XS,YS,ZS,MSGEN,CSHYET
      REAL RMCSH
C
```

```
C     Subroutine MCSHANT to move a diffusing CSH species and check for
C      reaction
C     Inputs: x,y, and z coordinates of current location of species
C     Returns: Flag (CSHYET) indicating if reaction has occurred
C
C      GENERATE MOVE
C
      RMCSH=RAN1(ISEED)
      MSGEN=6*RMCSH+1
      XS=XMCSH
      YS=YMCSH
      ZS=ZMCSH
      IF(MSGEN.EQ.1) THEN
        XS=XS-1
        IF(XS.LT.1) XS=SYSSIZE
      ENDIF
      IF(MSGEN.EQ.2) THEN
        XS=XS+1
        IF(XS.GT.SYSSIZE) XS=1
      ENDIF
      IF(MSGEN.EQ.3) THEN
        YS=YS-1
        IF(YS.LT.1) YS=SYSSIZE
      ENDIF
      IF(MSGEN.EQ.4) THEN
        YS=YS+1
      IF(YS.GT.SYSSIZE) YS=1
      ENDIF
      IF(MSGEN.EQ.5) THEN
        ZS=ZS-1
        IF(ZS.LT.1) ZS=SYSSIZE
      ENDIF
      IF(MSGEN.EQ.6) THEN
        ZS=ZS+1
        IF(ZS.GT.SYSSIZE) ZS=1
      ENDIF
C
C     If diffusing CSH encounters C3S or solid CSH, it is
C      converted to solid CSH
C
      IF((CEMENT(XS,YS,ZS).EQ.1).OR.(CEMENT(XS,YS,ZS).EQ.4).
     &OR.(NTIMES.EQ.CYCLENO)) THEN
        CEMENT(XMCSH,YMCSH,ZMCSH)=4
        CSHYET=0
      ENDIF
      IF((CEMENT(XS,YS,ZS).EQ.0).AND.(NTIMES.NE.CYCLENO)) THEN
        CEMENT(XS,YS,ZS)=3
        CEMENT(XMCSH,YMCSH,ZMCSH)=0
        XMCSH=XS
        YMCSH=YS
        ZMCSH=ZS
      ENDIF
      RETURN
      END
      SUBROUTINE MEASURE
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     +AGGSIZE,NCEMENT,NCEMDIS
      INTEGER NPOR,NCSH,NC3S,NCH,NINERT,NPOZZ,NAGG
C
C      Subroutine MEASURE to assess phase fractions in 3-D system
C      Called by: MAIN program
C
C      Initialize counters for various phases
C
      NPOR=0
      NCSH=0
      NCH=0
      NC3S=0
      NINERT=0
      NPOZZ=0
      NAGG=0
C
C      Update counters for all locations in 3-D system
C
      DO 14 I=1,SYSSIZE
      DO 14 J=1,SYSSIZE
      DO 14 K=1,SYSSIZE
      IF(CEMENT(I,J,K).EQ.0) NPOR=NPOR+1
      IF(CEMENT(I,J,K).EQ.4) NCSH=NCSH+1
      IF(CEMENT(I,J,K).EQ.1) NC3S=NC3S+1
      IF(CEMENT(I,J,K).EQ.9) NINERT=NINERT+1
      IF(CEMENT(I,J,K).EQ.10) NPOZZ=NPOZZ+1
      IF(CEMENT(I,J,K).EQ.5) NCH=NCH+1
      IF(CEMENT(I,J,K).EQ.8) NAGG=NAGG+1
   14 CONTINUE
```

```
C
C     Output Results
C
      WRITE(6,*)'Porosity=  ',NPOR
      WRITE(6,*)'C3S=  ',NC3S
      WRITE(6,*)'C-S-H=  ',NCSH
      WRITE(6,*)'CH=  ',NCH
      WRITE(6,*)'Inert=  ',NINERT
      WRITE(6,*)'Pozzolanic=  ',NPOZZ
      WRITE(6,*)'Aggregate=  ',NAGG
      RETURN
      END
      SUBROUTINE MEASAGG
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     +AGGSIZE,NCEMENT,NCEMDIS
      INTEGER PHASE(11),PTOT,PHREAD,IDIST,ICNT,IXL,IXR,IY,IZ
C
C     Subroutine MEASAGG to measure phase fractions as a function of
C     distance from aggregate surface
C     Called by: MAIN program
C
      WRITE(6,*)'Distance  Porosity  C3S  C-S-H  CH  Inert  Pozz'
C
C     Measure phase fractions as distance from aggregate increases
C
      DO 61 IDIST=1,((SYSSIZE-AGGSIZE)/2)
C
C      Initialize phase counts for this distance
C
      PTOT=0
      DO 62 ICNT=1,11
   62 PHASE(ICNT)=0
C
C     Check all pixels which are this distance from aggregate
C
        IXL=((SYSSIZE-AGGSIZE+2)/2)-IDIST
        IXR=((SYSSIZE+AGGSIZE)/2)+IDIST
      DO 63 IY=1,SYSSIZE
      DO 63 IZ=1,SYSSIZE
C
C     Check pixel left of aggregate
```

```
C
        PHID=1+CEMENT(IXL,IY,IZ)
        PHASE(PHID)=PHASE(PHID)+1
        PTOT=PTOT+1
C
C     Check pixel right of aggregate
C
        PHID=1+CEMENT(IXR,IY,IZ)
        PHASE(PHID)=PHASE(PHID)+1
 63     PTOT=PTOT+1
      WRITE(6,*)IDIST,PHASE(1),PHASE(2),PHASE(5),PHASE(6),
     +PHASE(10),PHASE(11)
  61  CONTINUE
      RETURN
      END
      SUBROUTINE CONNECT
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     +AGGSIZE,NCEMENT,NCEMDIS
      INTEGER NTOP,NTHROUGH,NCUR,NMATX(8000),NMATY(8000)
      INTEGER NMATZ(8000),XCN,YCN,ZCN,X1,Y1,Z1,IGOOD,NNEW
      INTEGER NTOT,NNEWX(8000),NNEWY(8000),NNEWZ(8000)
      INTEGER NPIX
C
C     Subroutine CONNECT to assess connectivity (percolation) of
C     a single phase
C
      WRITE(6,*)'ENTER PHASE TO ANALYZE 0) PORES 1) C3S'
      WRITE(6,*)'  4) CSH  5) CH  9) Inert  10) Pozzolanic'
      READ(5,*)NPIX
      WRITE(6,*)NPIX
C
C     Counters for number of pixels of phase accessible from
C     top and number which are part of a percolation path
C
      NTOP=0
      NTHROUGH=0
C
C     Percolation is assessed from top (K=1) to bottom (K=SYSSIZE)
C
      K=1
C
```

```
C     Start a burn pattern from all pixels on the top surface
C
      DO 17 I=1,SYSSIZE
      DO 17 J=1,SYSSIZE
      NCUR=0
      NTOT=0
      IGOOD=0
      IF(CEMENT(I,J,K).EQ.NPIX) THEN
C
C     Use a temporary ID of 7 to identify burnt pixels
C
       CEMENT(I,J,K)=7
        NTOT=NTOT+1
        NCUR=NCUR+1
C
C     Store the burn front coordinates in the matrices NMAT* and
C     NNEW*
C
        NMATX(NCUR)=I
        NMATY(NCUR)=J
        NMATZ(NCUR)=1
 57      NNEW=0
C
C     Propagate fire from all pixels in the current burn front
C
        DO 27 INEW=1,NCUR
          XCN=NMATX(INEW)
          YCN=NMATY(INEW)
          ZCN=NMATZ(INEW)
C
C      Check for propagation in all six directions
C
          DO 37 JNEW=1,6
            X1=XCN
            Y1=YCN
            Z1=ZCN
            IF(JNEW.EQ.1) X1=XCN-1
            IF(JNEW.EQ.2) X1=XCN+1
            IF(JNEW.EQ.3) Y1=YCN-1
            IF(JNEW.EQ.4) Y1=YCN+1
            IF(JNEW.EQ.5) Z1=ZCN-1
            IF(JNEW.EQ.6) Z1=ZCN+1
C
C      Note that burning is non-periodic
```

```
C
      IF((X1.GE.1).AND.(X1.LE.SYSSIZE).AND.(Y1.GE.1).
     &AND.(Y1.LE.SYSSIZE).AND.(Z1.GE.1).AND.(Z1.LE.SYSSIZE)) THEN
       IF(CEMENT(X1,Y1,Z1).EQ.NPIX) THEN
         NTOT=NTOT+1
         CEMENT(X1,Y1,Z1)=7
         NNEW=NNEW+1
         NNEWX(NNEW)=X1
         NNEWY(NNEW)=Y1
         NNEWZ(NNEW)=Z1
C
C        Check if new burnt pixel is on the bottom face of the system
C
         IF(Z1.EQ.SYSSIZE) THEN
           IGOOD=1
        ENDIF
       ENDIF
     ENDIF
  37  CONTINUE
  27  CONTINUE
      IF(NNEW.GT.0) THEN
       NCUR=NNEW
C
C      Copy the new burn front locations to matrices NMAT*
C
       DO 47 ICUR=1,NCUR
         NMATX(ICUR)=NNEWX(ICUR)
         NMATY(ICUR)=NNEWY(ICUR)
         NMATZ(ICUR)=NNEWZ(ICUR)
  47  CONTINUE
      GOTO 57
      ENDIF
      NTOP=NTOP+NTOT
      IF(IGOOD.EQ.1) THEN
       NTHROUGH=NTHROUGH+NTOT
      ENDIF
      ENDIF
  17  CONTINUE
      WRITE(6,*)'Phase ID= ',NPIX
      WRITE(6,*)'Number accessible from top= ',NTOP
      WRITE(6,*)'Number contained in through pathways= ',NTHROUGH
C
C     Restore all burnt pixels to original phase IDs
C
```

```
      DO 67 I=1,SYSSIZE
      DO 67 J=1,SYSSIZE
      DO 67 K=1,SYSSIZE
      IF(CEMENT(I,J,K).EQ.7)  CEMENT(I,J,K)=NPIX
   67 CONTINUE
      RETURN
      END
      SUBROUTINE CONSOLD
      INTEGER CEMENT(101,101,101),CYCLENO,NTIMES,NFILL,NPZR
      INTEGER SYSSIZE,AGGSIZE,NCEMENT,NCEMDIS
      INTEGER*4 ISEED
      COMMON /A/CEMENT,CYCLENO,NTIMES,NFILL,NPZR,ISEED,SYSSIZE,
     +AGGSIZE,NCEMENT,NCEMDIS
      INTEGER NTOP,NTHROUGH,NCUR,NMATX(8000),NMATY(8000)
      INTEGER NMATZ(8000),XCN,YCN,ZCN,X1,Y1,Z1,IGOOD,NNEW
      INTEGER NTOT,NNEWX(8000),NNEWY(8000),NNEWZ(8000)
C
C      Subroutine CONSOLD to assess connectivity (percolation) of
C      total solid phases with exception of aggregate if
C      one is present
C
C
C      Counters for number of pixels of solid accessible from
C      top and number which are part of a percolation path
C
      NTOP=0
      NTHROUGH=0
C
C      Percolation is assessed from top (K=1) to bottom (K=SYSSIZE)
C
      K=1
C
C      Start a burn pattern from all pixels on the top surface
C
      DO 17 I=1,SYSSIZE
      DO 17 J=1,SYSSIZE
      NCUR=0
      NTOT=0
      IGOOD=0
      IF((CEMENT(I,J,K).NE.0).AND.(CEMENT(I,J,K).LT.12)
     +.AND.(CEMENT(I,J,K).NE.8)) THEN
C
C      Use a temporary ID of 12 + present ID to identify burnt pixels
C
```

```
      CEMENT(I,J,K)=CEMENT(I,J,K)+12
       NTOT=NTOT+1
       NCUR=NCUR+1
C
C     Store the burn front coordinates in the matrices NMAT* and
C     NNEW*
C
       NMATX(NCUR)=I
       NMATY(NCUR)=J
       NMATZ(NCUR)=1
  57    NNEW=0
C
C     Propagate fire from all pixels in the current burn front
C
       DO 27 INEW=1,NCUR
         XCN=NMATX(INEW)
         YCN=NMATY(INEW)
         ZCN=NMATZ(INEW)
C
C      Check for propagation in all six directions
C
         DO 37 JNEW=1,6
           X1=XCN
           Y1=YCN
           Z1=ZCN
           IF(JNEW.EQ.1) X1=XCN-1
           IF(JNEW.EQ.2) X1=XCN+1
           IF(JNEW.EQ.3) Y1=YCN-1
           IF(JNEW.EQ.4) Y1=YCN+1
           IF(JNEW.EQ.5) Z1=ZCN-1
           IF(JNEW.EQ.6) Z1=ZCN+1
C
C      Note that burning is non-periodic
C
      IF((X1.GE.1).AND.(X1.LE.SYSSIZE).AND.(Y1.GE.1).
     &AND.(Y1.LE.SYSSIZE).AND.(Z1.GE.1).AND.(Z1.LE.SYSSIZE)) THEN
      IF((CEMENT(X1,Y1,Z1).LT.12).AND.(CEMENT(X1,Y1,Z1).NE.0)
     +.AND.(CEMENT(X1,Y1,Z1).NE.8)) THEN
         NTOT=NTOT+1
         CEMENT(X1,Y1,Z1)=CEMENT(X1,Y1,Z1)+12
         NNEW=NNEW+1
         NNEWX(NNEW)=X1
         NNEWY(NNEW)=Y1
         NNEWZ(NNEW)=Z1
```

```
C
C       Check if new burnt pixel is on the bottom face of the system
C
         IF(Z1.EQ.SYSSIZE) THEN
           IGOOD=1
         ENDIF
        ENDIF
      ENDIF
  37  CONTINUE
  27  CONTINUE
       IF(NNEW.GT.0) THEN
        NCUR=NNEW
C
C       Copy the new burn front locations to matrices NMAT*
C
        DO 47 ICUR=1,NCUR
          NMATX(ICUR)=NNEWX(ICUR)
          NMATY(ICUR)=NNEWY(ICUR)
          NMATZ(ICUR)=NNEWZ(ICUR)
  47  CONTINUE
      GOTO 57
      ENDIF
      NTOP=NTOP+NTOT
      IF(IGOOD.EQ.1) THEN
        NTHROUGH=NTHROUGH+NTOT
      ENDIF
      ENDIF
  17  CONTINUE
      WRITE(6,*)'FOR TOTAL SOLIDS '
      WRITE(6,*)'Number accessible from top=  ',NTOP
      WRITE(6,*)'Number contained in through pathways=  ',NTHROUGH
C
C      Restore all burnt pixels to original phase IDs
C
      DO 67 I=1,SYSSIZE
      DO 67 J=1,SYSSIZE
      DO 67 K=1,SYSSIZE
      IF(CEMENT(I,J,K).GT.12)  CEMENT(I,J,K)=CEMENT(I,J,K)-12
  67  CONTINUE
      RETURN
      END
```

APPENDIX C
C Listing for HYDRA3D

```
#include <stdio.h>
#include <math.h>

#define SYSSIZE 100

/* phase identifiers */
#define POROSITY 0
#define C3S 1
#define DIFFCH 2
#define DIFFCSH 3
#define CSH 4
#define CH 5
#define SURFID 6
#define BURNT 7
#define AGG 8
#define INERT 9
#define POZZ 10

/* number of different classes of spheres allowed */
#define NUMSIZES 10

/* definitions for portable random number generator */
#define M1 259200
#define IA1 7141
#define IC1 54773
#define RM1 (1.0/M1)
#define M2 134456
#define IA2 8121
#define IC2 28411
#define RM2 (1.0/M2)
#define M3 243000
#define IA3 4561
#define IC3 51349


/* 3-D microstructure is stored in 3-D array cement */
static unsigned short int cement [101] [101] [101];
long int nfill,npr,ncement,ncemdis;
int cycleno,ntimes,aggsize,*seed;
```

```
/*########################## ####################################*/
/*                                                                */
/*      Program:HYDRA3D.C                                         */
/*      Programmer: Dale P. Bentz                                 */
/*              Building and Fire Research Laboratory             */
/*              National Institute of Standards and Technology    */
/*              Building 226  Room B-348                          */
/*              Gaithersburg, MD  20899-0001                      */
/*              (301) 975-5865  FAX (301) 975-4032                */
/*      Date: 8/91                                                */
/*                                                                */
/*      Purpose: To execute a three-dimensional cement            */
/*              hydration model                                   */
/*                                                                */
/*########################## ####################################*/

/* Portable random number generator routine, ran1, from */
/* Numerical Recipes in C */
/* Press, Flannery, Teukolsky, and Vetterling */
/* Returns floating point random numbers between 0 and 1 */
float ran1(idum)
int *idum;
{
        static long ix1,ix2,ix3;
        static float r[98];
        float temp;
        static int iff=0;
        int j;

        if (*idum < 0 || iff == 0) {
                iff=1;
                ix1=(IC1-(*idum)) % M1;
                ix1=(IA1*ix1+IC1) % M1;
                ix2=ix1 % M2;
                ix1=(IA1*ix1+IC1) % M1;
                ix3=ix1 % M3;
                for (j=1;j<=97;j++) {
                        ix1=(IA1*ix1+IC1) % M1;
                        ix2=(IA2*ix2+IC2) % M2;
                        r[j]=(ix1+ix2*RM2)*RM1;
                }
                *idum=1;
        }
        ix1=(IA1*ix1+IC1) % M1;
        ix2=(IA2*ix2+IC2) % M2;
        ix3=(IA3*ix3+IC3) % M3;
        j=1 + ((97*ix3)/M3);
        if (j > 97 || j < 1) printf("RAN1: This cannot happen.");
        temp=r[j];
        r[j]=(ix1+ix2*RM2)*RM1;
        return temp;
}

/* undefine variables in case needed later */
#undef M1
#undef IA1
#undef IC1
#undef RM1
#undef M2
#undef IA2
#undef IC2
#undef RM2
#undef M3
#undef IA3
#undef IC3

/* routine to display an introduction to the user */
void intro()
{
        printf("Welcome to HYDRA3D, a digital-image-based cement \n");
        printf("microstructural model.  This program has been \n");
        printf("developed to simulate the microstructural \n");
        printf("development of cement, specifically tricalcium \n");
        printf("silicate, C3S, as it reacts with water.  In \n");
        printf("addition to C3S, the user may also add a single \n");
        printf("inert aggregate and/or inert or pozzolanic mineral \n");
        printf("admixture particles to the microstructure.  In \n");
        printf("addition to hydration, the user may also assess \n");
        printf("phase fractions (either globally or as a function \n");
        printf("of distance from the aggregate surface) and \n");
        printf("the connectivity of any individual phase or of \n");
        printf("the total solids (neglecting any aggregate) present \n");
        printf("in the system. \n \n");

}

/* Routine to add a flat plate aggregate through the 3-D microstructure */
void addagg()
```

```
{
        int ix,iy,iz;

        printf("Enter aggregate thickness (even number of pixels) \n");
        scanf("%d",&aggsize);
        printf("%d \n",aggsize);

/* Aggregate extends through entire system in y and z directions */
        for(ix=((SYSSIZE-aggsize+2)/2);ix<=((SYSSIZE+aggsize)/2);ix++){
        for(iy=1;iy<=SYSSIZE;iy++){
        for(iz=1;iz<=SYSSIZE;iz++){
                cement [ix] [iy] [iz]=AGG;
        }
        }
        }

}

/* routine to check or perform placement of sphere centered */
/* at location xin,yin,zin of radius radd */
/* wflg=1 check for fit of sphere */
/* wflg=2 place the sphere */
int chksph(xin,yin,zin,radd,wflg,phasein)
        int xin,yin,zin,radd,wflg,phasein;
{
        int sflg,nofits,xp,yp,zp,i,j,k;
        float dist;

        nofits=0;    /* Flag indicating if placement is possible */

/* Check all pixels within the digitized sphere volume */
        for(i=xin-radd;((i<=xin+radd)&&(nofits==0));i++){
        for(j=yin-radd;((j<=yin+radd)&&(nofits==0));j++){
        for(k=zin-radd;((k<=zin+radd)&&(nofits==0));k++){

                xp=i;
                yp=j;
                zp=k;
                /* use periodic boundary conditions for sphere placement */
                if(xp<1) {xp+=SYSSIZE;}
                if(yp<1) {yp+=SYSSIZE;}
                if(zp<1) {zp+=SYSSIZE;}
                if(xp>SYSSIZE) {xp-=SYSSIZE;}
                if(yp>SYSSIZE) {yp-=SYSSIZE;}
                if(zp>SYSSIZE) {zp-=SYSSIZE;}

        /* Compute distance from center of sphere to this pixel */
                dist=sqrt((float)((i-xin)*(i-xin)+(j-yin)*(j-yin)+(k-zin)*(k-zin)));
                if((dist-0.5)<=(float)radd){
                                if(wflg==2){
                                        cement [xp] [yp] [zp] =phasein;
        /* Update counter of C3S species if necessary */
                                        if(phasein==C3S){
                                                ncement+=1;
                                        }
        /* Update counter of pozzolanic filler species if necessary */
                                        if(phasein==POZZ){
                                                nfill+=1;
                                        }
                                }
                        if((wflg==1)&&(cement [xp] [yp] [zp] !=POROSITY)){
                                        nofits=1;
                        }
                }
        }
        }
        }

        /* return flag indicating if sphere will fit */
        return(nofits);
}

/* routine to place spheres of various sizes and phases at random */
/* locations in 3-D microstructure */
void gsphere(numgen,numeach,sizeeach,pheach)
        int numgen;
        long int numeach[NUMSIZES];
        int sizeeach[NUMSIZES],pheach[NUMSIZES];
{
        int count,x,y,z,radius,ig,kg,phsph;
        long int jg;
        float rx,ry,rz;


/* Generate spheres of each size class in turn (largest first) */
        for(ig=0;ig<numgen;ig++){

                radius=sizeeach[ig];        /* radius for this class */
```

```
		phsph=pheach[ig];		/* phase for this class */
		/* loop for each sphere in this size class */
		for(jg=1;jg<=numeach[ig];jg++){

			do{
			/* generate a random center location for the sphere */
				rx=ran1(seed);
				ry=ran1(seed);
				rz=ran1(seed);
				x=(int)((float)SYSSIZE*rx)+1;
				y=(int)((float)SYSSIZE*ry)+1;
				z=(int)((float)SYSSIZE*rz)+1;
				/* see if the sphere will fit at x,y,z */
				count=chksph(x,y,z,radius,1,phsph);
			} while(count!=0);

			/* place the sphere at x,y,z */
			count=chksph(x,y,z,radius,2,phsph);
		}
	}
}

/* routine to obtain user input and create a starting microstructure */
void create()
{
	int numsize,sphrad [NUMSIZES],sphid [NUMSIZES];
	long int sphnum [NUMSIZES],inval1;
	int isph,inval;

	printf("Enter number of different size spheres to use (maximum is 10) \n");
	scanf("%d",&numsize);
	printf("%d \n",numsize);

	if((numsize>0)&&(numsize<(NUMSIZES+1))){
		printf("Enter number, size, and phase ID for each class (largest radius
1st) \n");

	/* Obtain input for each size class of spheres */
		for(isph=0;isph<numsize;isph++){
			printf("Enter number of spheres of class %d \n",isph+1);
			scanf("%ld",&inval1);
			printf("%ld \n",inval1);
			sphnum[isph]=inval1;
```

```
                printf("Enter radius of spheres of class %d \n",isph+1);
                printf("(Integer <=10 please) \n");
                scanf("%d",&inval);
                printf("%d \n",inval);
                sphrad[isph]=inval;

                printf("Enter phase to create for spheres of class %d \n",isph+1);
                printf("(1-C3S, 9- Inert filler, 10- Pozzolanic filler \n");
                scanf("%d",&inval);
                printf("%d \n",inval);
                sphid[isph]=inval;
            }
            gsphere(numsize,sphnum,sphrad,sphid);
        }
}

/* routine to place one pixel filler particles at random
   unoccupied locations in 3-D system                    */
void genfill(ntopl,idtouse)
        long int ntopl;  /* Number of filler pixels to place */
        int idtouse;  /* Phase to be assigned to filler */
{
        int pfill,xfill,yfill,zfill;
        long int ifill;
        float rxf,ryf,rzf;

/* Place each filler particle in turn */
        for(ifill=1;ifill<=ntopl;ifill++){

            pfill=0;
            /* place this filler pixel at a random unoccupied location */
            while (pfill==0){
                    rxf=ran1(seed);
                    ryf=ran1(seed);
                    rzf=ran1(seed);

                    xfill=(int)((float)SYSSIZE*rxf)+1;
                    yfill=(int)((float)SYSSIZE*ryf)+1;
                    zfill=(int)((float)SYSSIZE*rzf)+1;

                    if(cement [xfill] [yfill] [zfill] ==POROSITY){
                        pfill=1;
                        cement [xfill] [yfill] [zfill]=idtouse;
                    }
```

```
				}
		}
}

/* subroutine to obtain user input as to filler to be added */
void filler()
{
		long int nadd;
		int fillid;

		printf("Enter number of filler particles to add \n");
		scanf("%ld",&nadd);
		printf("%ld \n",nadd);
		printf("Enter filled id to use (9- inert, 10- pozzolanic) \n");
		scanf("%d",&fillid);
		printf("%d \n",fillid);

/* If phase ID is valid, call routine to place the filler */
		if((fillid= =INERT)||(fillid= =POZZ)){
				genfill(nadd,fillid);
				if(fillid= =POZZ){
						nfill+ =nadd;
				}
		}
}

/* routine to perform the dissolution process for a hydration cycle */
/* Locations of dissolved species are returned in arrays disx,disy, and */
/* disz                                                                  */
long int dissolv(disx,disy,disz)
		unsigned short int disx [200000], disy [200000], disz [200000];
{
		long int numdis,nchadd,ncshadd,numorg,i,j,k;
		int xc,yc,zc,irnd,nedge;
		float rdis,rdx,rdy,rdz;

/* counter for number of species dissolved */
		numdis=0;

/*		Pass 1: Highlight all edge points		*/
/*				(C3S with at least one neighbor porosity) */

		for(i=1;i< =SYSSIZE;i+ +){
		for(j=1;j< =SYSSIZE;j+ +){
```

```
	for(k=1;k<=SYSSIZE;k++){
		/* if pixel is cement, see if it is on an edge */
		if(cement [i] [j] [k] ==C3S){
			nedge=1;
			xc=i-1;
			/* periodic boundaries once more */
			if(xc<1) {xc=SYSSIZE;}
			nedge*=cement [xc] [j] [k];
			xc=i+1;
			if(xc>SYSSIZE) {xc=1;}
			nedge*=cement [xc] [j] [k];
			yc=j-1;
			if(yc<1) {yc=SYSSIZE;}
			nedge*=cement [i] [yc] [k];
			yc=j+1;
			if(yc>SYSSIZE) {yc=1;}
			nedge*=cement [i] [yc] [k];
			zc=k-1;
			if(zc<1) {zc=SYSSIZE;}
			nedge*=cement [i] [j] [zc];
			zc=k+1;
			if(zc>SYSSIZE) {zc=1;}
			nedge*=cement [i] [j] [zc];
		/* if nedge is zero, at least one neighbor was porosity */
			if(nedge==0){cement [i] [j] [k]=SURFID;}
		}
	}
	}
	}

/*	Pass 2: Dissolve all highlighted edge points at random */
/*		by allowing each to attempt a one step random walk */

	for(i=1;i<=SYSSIZE;i++){
	for(j=1;j<=SYSSIZE;j++){
	for(k=1;k<=SYSSIZE;k++){
		if(cement [i] [j] [k]==SURFID){
		/* Choose a random direction for the step */
			rdis=ran1(seed);
			irnd=(int)(6.*rdis)+1;
			xc=i;
			yc=j;
			zc=k;
			/* dissolution is attempted by performing */
```

```
        /* a one-step random walk */
        switch (irnd) {
                    case 1:
                            xc-=1;
                            if(xc<1) {xc=SYSSIZE;}
                            break;
                    case 2:
                            xc+=1;
                            if(xc>SYSSIZE) {xc=1;}
                            break;
                    case 3:
                            yc-=1;
                            if(yc<1) {yc=SYSSIZE;}
                            break;
                    case 4:
                            yc+=1;
                            if(yc>SYSSIZE) {yc=1;}
                            break;
                    case 5:
                            zc-=1;
                            if(zc<1) {zc=SYSSIZE;}
                            break;
                    case 6:
                            zc+=1;
                            if(zc>SYSSIZE) {zc=1;}
                            break;
                    default:
                            break;
                }
/* if step is not into porosity, remain as solid C3S */
        if(cement [xc] [yc] [zc] !=POROSITY){
                cement [i] [j] [k]=C3S;
        }
        /* if step is into porosity, perform the dissolution */
        /* and store location of dissolved species */
        if(cement [xc] [yc] [zc]==POROSITY){
                cement [xc] [yc] [zc]=DIFFCSH;
                cement [i] [j] [k]=POROSITY;
                numdis+=1;
                disx[numdis]=xc;
                disy[numdis]=yc;
                disz[numdis]=zc;
                }
}
```

```
        }
        }
        }

        printf("Original number species dissolved=  %ld \n",numdis);
        numorg=numdis;
        ncemdis+ =numdis;

/*      Add in extra diffusing species */
/* One dissolved unit of C3S should produce 1.7 units of C-S-H */
/* and 0.61 units of CH */
/* Expansion factors of 0.7 and 0.61 are taken from work of */
/* Young & Hansen, MRS Proceedings, Vol. 85 , pp. 313-322, 1987    */
        ncshadd=(int)((float)numdis*0.7);
        nchadd=(int)((float)numdis*0.61);
        numdis+ =(nchadd+ncshadd);
        if(numdis> =200000){
                printf("Too many dissolved species generated \n");
                printf("Aborting run \n");
                exit(1);
        }

        for(i=1;i< =(nchadd+ncshadd);i+ +){
                nedge=0;    /* flag indicating successful placement */
                do{
                /* extra diffusing species are added at totally */
                /* random unoccupied locations in system */
                        rdx=ran1(seed);
                        rdy=ran1(seed);
                        rdz=ran1(seed);
                        xc=(int)((float)SYSSIZE*rdx)+1;
                        yc=(int)((float)SYSSIZE*rdy)+1;
                        zc=(int)((float)SYSSIZE*rdz)+1;

                        if(cement [xc] [yc] [zc]= =POROSITY){
                                nedge=1;
                                disx[numorg+i]=xc;
                                disy[numorg+i]=yc;
                                disz[numorg+i]=zc;

                                if(i< =nchadd){
                                        cement [xc] [yc] [zc]=DIFFCH;
                                }
                                if(i>nchadd){
                                        cement [xc] [yc] [zc]=DIFFCSH;
                                }
                        }
                } while (nedge= =0);
        }

/* return number of generated diffusing species */
        return(numdis);
}

/* routine to add in extra pozzolanic CSH when diffusing CH */
/* species reacts with pozzolanic filler */
void addext(xinn,yinn,zinn)
        int xinn,yinn,zinn;  /* location of reaction site */
{
        int xchr,ychr,zchr,fchr,mgen1;
        long int tfact;        /* indicates when all 6 neighbors have been tested */
        float rmadd;

/*      Add in extra pozzolanic CSH to account for volume balance */
/*      Random tries at neighboring locations (6) until all tried */
        fchr=0;       /* Flag indicating successful placement */
        tfact=1;
        while((tfact!=30030)&&(fchr= =0)){
        /* Generate a random direction to test */
                rmadd=ran1(seed);
                mgen1=(int)(6.*rmadd)+1;
                xchr=xinn;
                ychr=yinn;
                zchr=zinn;

                switch(mgen1){
                        case 1:
                                xchr-=1;
                                /* periodic boundaries once again */
                                if(xchr<1) {xchr=SYSSIZE;}
                                if((tfact%2)!=0){tfact*=2;}
                                break;
                        case 2:
                                xchr+=1;
                                if(xchr>SYSSIZE) {xchr=1;}
                                if((tfact%3)!=0){tfact*=3;}
                                break;
                        case 3:
```

```
                                        ychr-=1;
                                        if(ychr<1) {ychr=SYSSIZE;}
                                        if((tfact%5)!=0){tfact*=5;}
                                        break;
                                case 4:
                                        ychr+=1;
                                        if(ychr>SYSSIZE) {ychr=1;}
                                        if((tfact%7)!=0){tfact*=7;}
                                        break;
                                case 5:
                                        zchr-=1;
                                        if(zchr<1) {zchr=SYSSIZE;}
                                        if((tfact%11)!=0){tfact*=11;}
                                        break;
                                case 6:
                                        zchr+=1;
                                        if(zchr>SYSSIZE) {zchr=1;}
                                        if((tfact%13)!=0){tfact*=13;}
                                        break;
                                default:
                                        break;
                        }
                if(cement [xchr] [ychr] [zchr]==POROSITY){
                        cement [xchr] [ychr] [zchr]=POZZ;
                        fchr=1;
                }
        }

/*      If initial efforts unsuccessful, then */
/*      Add extra CSH at random location in pore space */

        while(fchr==0){
                        rmadd=ran1(seed);
                        xchr=(int)((float)SYSSIZE*rmadd)+1;
                        rmadd=ran1(seed);
                        ychr=(int)((float)SYSSIZE*rmadd)+1;
                        rmadd=ran1(seed);
                        zchr=(int)((float)SYSSIZE*rmadd)+1;
                        if(cement [xchr] [ychr] [zchr]==POROSITY){
                                cement [xchr] [ychr] [zchr]=POZZ;
                                fchr=1;
                        }
        }
}
```

```
/* routine to move/react a diffusing CH species */
int mvchant(xmch1,ymch1,zmch1,pnuc)
        int *xmch1,*ymch1,*zmch1;    /* location of CH species to move */
        float pnuc;
{
        int xm,ym,zm,mgen,chyet,xmch,ymch,zmch;
        float pgen,rm;

        zmch=(*zmch1);
        xmch=(*xmch1);
        ymch=(*ymch1);
        chyet=1;
        /* first allow for random nucleation at current location */
        /* if last step in this cycle, convert to solid CH */
        pgen=ran1(seed);
        if((pnuc>pgen)||(cycleno==ntimes)){
                cement [xmch] [ymch] [zmch]=CH;
                chyet=0;
        }

        /* If no nucleation, allow for diffusion */
        if(chyet==1){

/*      Generate a move */
                rm=ran1(seed);
                mgen=(int)(6.*rm)+1;
                xm=xmch;
                ym=ymch;
                zm=zmch;
                switch (mgen){
                                case 1:
                                        xm-=1;
                                        if(xm<1) {xm=SYSSIZE;}
                                        break;
                                case 2:
                                        xm+=1;
                                        if(xm>SYSSIZE) {xm=1;}
                                        break;
                                case 3:
                                        ym-=1;
                                        if(ym<1) {ym=SYSSIZE;}
                                        break;
                                case 4:
```

```
                        ym+ =1;
                        if(ym>SYSSIZE) {ym=1;}
                        break;
                case 5:
                        zm- =1;
                        if(zm<1) {zm=SYSSIZE;}
                        break;
                case 6:
                        zm+ =1;
                        if(zm>SYSSIZE) {zm=1;}
                        break;
                default:
                        break;
}
/* check for growth of a solid CH crystal */
if(cement [xm] [ym] [zm]= =CH){
        cement [xmch] [ymch] [zmch]=CH;
        chyet=0;
}
/* check for pozzolanic reaction of CH */
/* Each volume unit of pozzolanic filler can */
/* react with 2.08 volume units of CH to produce */
/* 4.6 volume untis of pozzolanic C-S-H */
/* This assumes the pozzolanic filler is pure silica */
/* with a molar volume of 27 cm^3/mole */
if((cement [xm] [ym] [zm]= =POZZ)&&
  (npr< =(int)((float)nfill*2.08))){
        cement [xmch] [ymch] [zmch]=POZZ;
        chyet=0;
        npr+ =1;
        rm=ran1(seed);
        /* Expansion probability = (4.6-1)/2.08-1=0.73 */
        /* where the 1 taken away from the 4.6 represents */
        /* the original silica fume particle pixel */
        if(rm< =0.73){
                addext(xmch,ymch,zmch);
        }
}
if(cement [xm] [ym] [zm]= =POROSITY){
/* Diffusion by moving species to new location */
        cement [xm] [ym] [zm]=DIFFCH;
        cement [xmch] [ymch] [zmch]=POROSITY;
        xmch=xm;
        ymch=ym;
```

```
                    zmch=zm;
                }
        }
        *zmch1=zmch;
        *ymch1=ymch;
        *xmch1=xmch;

/* return flag indicating if diffusing CH species reacted */
        return(chyet);
}

/* routine to move/react a diffusing CSH species */
int mcshant(xmcsh1,ymcsh1,zmcsh1)
        int *xmcsh1,*ymcsh1,*zmcsh1;
{
        int xmcsh,ymcsh,zmcsh,xs,ys,zs,msgen,cshyet;
        float rmcsh;

        cshyet=1;
        /* choose a random direction for the move */
        rmcsh=ran1(seed);
        msgen=(int)(6.*rmcsh)+1;
        xmcsh=(*xmcsh1);
        ymcsh=(*ymcsh1);
        zmcsh=(*zmcsh1);

        xs=xmcsh;
        ys=ymcsh;
        zs=zmcsh;

        switch(msgen) {
                case 1:
                        xs=xs-1;
                        if(xs<1) {xs=SYSSIZE;}
                        break;
                case 2:
                        xs+=1;
                        if(xs>SYSSIZE) {xs=1;}
                        break;
                case 3:
                        ys-=1;
                        if(ys<1) {ys=SYSSIZE;}
                        break;
                case 4:
                        ys+=1;
                        if(ys>SYSSIZE) {ys=1;}
                        break;
                case 5:
                        zs-=1;
                        if(zs<1) {zs=SYSSIZE;}
                        break;
                case 6:
                        zs+=1;
                        if(zs>SYSSIZE) {zs=1;}
                        break;
                default:
                        break;
        }

        /* check for reaction at solid C3S or C-S-H surface */
        /* If last diffusion step this cycle, convert to solid C-S-H */
        if((cement [xs] [ys] [zs]==C3S)||(cement [xs] [ys] [zs]==CSH)
          ||(ntimes==cycleno)){
                cement [xmcsh] [ymcsh] [zmcsh]=CSH;
                cshyet=0;
        }
        if((cshyet!=0)&&(cement [xs] [ys] [zs]==POROSITY)){
        /* Diffusion by moving species to new location */
                cement [xs] [ys] [zs]=DIFFCSH;
                cement [xmcsh] [ymcsh] [zmcsh]=POROSITY;
                xmcsh=xs;
                ymcsh=ys;
                zmcsh=zs;
        }

        *xmcsh1=xmcsh;
        *ymcsh1=ymcsh;
        *zmcsh1=zmcsh;

/* return flag indicating if diffusing CSH species has reacted */
        return(cshyet);
}

/* routine to control the hydration process */
void hydrate()
{
        int niter,stopch;
        static unsigned short int antsz[200000],antsx[200000],antsy[200000];
```

```
int xant,yant,zant,alive;
int anttype,i,j,k;
long int numants,numleft,nbleft,nblue;
float nucscale,nucprob;
float beterm,bbias,bnew,almax,alpha;

do{
        printf("Do you wish to specify 0) max. # of cycles or 1) max.
                degree of hydration \n");
        scanf("%d",&stopch);
} while ((stopchl = 1)&&(stopchl = 0));
printf("%d \n",stopch);

if(stopch = = 0){
        printf("Enter number of hydration cycles (iterations) to perform \n");
        scanf("%d",&niter);
        printf("%d \n",niter);
        almax = 1.0;
}
if(stopch = = 1){
        printf("Enter maximum degree of hydration \n");
        scanf("%f",&almax);
        printf("%f \n",almax);
        niter = 5000;
}
printf("Enter maximum number of diffusion steps per cycle \n");
scanf("%d",&ntimes);
printf("%d \n",ntimes);

printf("Enter maximum probability for CH nucleation \n");
scanf("%f",&nucprob);
printf("%f \n",nucprob);
printf("Enter exponential scale factor for CH nucleation \n");
scanf("%f",&nucscale);
printf("%f \n",nucscale);

alpha = (float)ncemdis/(float)ncement;
for(i = 1;((i< = niter)&&(alpha<almax));i + + ){

        numants = 0;
        /* perform the dissolution step */
        numants = dissolv(antsx,antsy,antsz);
        numleft = numants;  /* Number of diffusing species remaining */
        alpha = (float)ncemdis/(float)ncement;
```

```
                printf("Number dissolved  =  %ld \n",numants);
                nblue=(int)(61.*(float)numants)/231.0;

/* diffuse until all species have reacted or maximum number of */
/* steps is exceeded */
                for(j=1;((j< =ntimes)&&(numleft!=0));j+ +){

                        cycleno=j;
                        numleft=0;
                        nbleft=0;
                        bnew=(float)nblue;
                        bnew/=nucscale;
                        beterm=exp(-bnew);
                        /* determine new probability of CH nucleation */
                        bbias=nucprob*(1.-beterm);

                /* Move each remaining diffusing species in turn */
                        for(k=1;k< =numants;k+ +){

                                /* get the current location and type */
                                /* of this diffusing species */
                                xant=antsx[k];
                                yant=antsy[k];
                                zant=antsz[k];
                                anttype=cement [xant] [yant] [zant];
                                alive=1;  /* Flag indicating reaction */

                /* Note that addresses are passed to routines mvchant and */
                /* mcshant so that new locations of diffusing species can */
                /* be updated (returned) */
                                if(anttype= =DIFFCH){
                                        alive=mvchant(&xant,&yant,&zant,bbias);
                                }
                                if(anttype= =DIFFCSH){
                                        alive=mcshant(&xant,&yant,&zant);
                                }
                                if(alive= =1){
                                        numleft+ =1;
                                        /* store the new location of the */
                                        /* diffusing species */
                        /* Note that we use only one array here, because */
                        /* the number of diffusing species remaining after */
                        /* a diffusion step is always less than the number */
                        /* present at the start of the step */
```

```
                                        antsx[numleft] = xant;
                                        antsy[numleft] = yant;
                                        antsz[numleft] = zant;

                                        if(anttype = = DIFFCH){
                                                nbleft + = 1;
                                        }
                                }
                        }

                        /* Update number of diffusing species still in system */
                        numants = numleft;
                        nblue = nbleft;
                }
        }
}

/* routine to assess global phase fractions present in 3-D system */
void measure()
{
        long int npor,ncsh,nc3s,nch,ninert,npozz,nagg;
        int i,j,k;

/* counters for the various phase fractions */
        npor = 0;
        ncsh = 0;
        nch = 0;
        nc3s = 0;
        npozz = 0;
        ninert = 0;
        nagg = 0;

/* Check all pixels in 3-D microstructure */
        for(i = 1;i < = SYSSIZE;i + +){
        for(j = 1;j < = SYSSIZE;j + +){
        for(k = 1;k < = SYSSIZE;k + +){

                if(cement [i] [j] [k] = = POROSITY) {npor + = 1;}
                if(cement [i] [j] [k] = = CSH) {ncsh + = 1;}
                if(cement [i] [j] [k] = = C3S) {nc3s + = 1;}
                if(cement [i] [j] [k] = = INERT) {ninert + = 1;}
                if(cement [i] [j] [k] = = POZZ) {npozz + = 1;}
                if(cement [i] [j] [k] = = CH) {nch + = 1;}
                if(cement [i] [j] [k] = = AGG) {nagg + = 1;}
        }
        }
        }

/* Output results */
        printf("Porosity=  %ld \n",npor);
        printf("C3S=  %ld \n",nc3s);
        printf("C-S-H=  %ld \n",ncsh);
        printf("CH=  %ld \n",nch);
        printf("Inert=  %ld \n",ninert);
        printf("Pozzolanic  =  %ld \n",npozz);
        printf("Aggregate=  %ld \n",nagg);

}

/* Routine to measure phase fractions as a function of distance from */
/* aggregate surface                                                  */
void measagg()
{
        int phase [11],ptot;
        int icnt,ix,iy,iz,phid,idist;

        printf("Distance  Porosity  C3S  C-S-H  CH  Inert  Pozzolanic \n");

/* Increase distance from aggregate in increments of one */
        for(idist=1;idist<=(SYSSIZE-aggsize)/2;idist++){

        /* Initialize phase counts for this distance */
                for(icnt=0;icnt<11;icnt++){
                        phase[icnt]=0;
                }
                ptot=0;

/* Check all pixels which are this distance from aggregate surface */
                for(iy=1;iy<=SYSSIZE;iy++){
                for(iz=1;iz<=SYSSIZE;iz++){
                        /* Pixel left of aggregate surface */
                        ix=((SYSSIZE-aggsize+2)/2)-idist;
                        phid=cement [ix] [iy] [iz];
                        ptot+=1;
                        phase[phid]+=1;

                        /* Pixel right of aggregate surface */
                        ix=((SYSSIZE+aggsize)/2)+idist;
                        phid=cement [ix] [iy] [iz];
                        ptot+ =1;
                        phase[phid]+ =1;
                }
                }

                /* Output results for this distance from surface */
                printf("%d   %d   %d  %d  %d   %d   %d \n", idist,phase[0],phase[1],
phase[4],phase[5],phase[9],phase[10]);

        }
}

/* routine to assess the connectivity (percolation) of a single phase */
/* Two matrices are used here: one to store the recently burnt locations */
/*                 the other to store the newly found burnt locations */
void connect()
{
        long int ntop,nthrough,ncur,nnew,ntot;
        int i,inew,j,k,nmatx[9000],nmaty[9000],nmatz[9000];
        int   xcn,ycn,zcn,npix,x1,y1,z1,igood,nnewx[9000],nnewy[9000],nnewz[9000];
        int jnew,icur;

        printf("Enter phase to analyze 0) pores 1) C3S 4) CSH 5) CH \n");
        printf("  9) Inert  10) Pozzolanic \n");
        scanf("%d",&npix);
        printf("%d \n",npix);

/* counters for number of pixels of phase accessible from top surface */
/* and number which are part of a percolated pathway */
        ntop=0;
        nthrough=0;

        /* percolation is assessed from top to bottom only */
        /* and burning algorithm is nonperiodic in x and y directions */
        k=1;

        for(i=1;i< =SYSSIZE;i+ +){
        for(j=1;j< =SYSSIZE;j+ +){

                ncur=0;
                ntot=0;
                igood=0;    /* Indicates if bottom has been reached */
                if(cement [i] [j] [k]= =npix){
                    /* Start a burn front */
                    cement [i] [j] [k]=BURNT;
                    ntot+ =1;
                    ncur+ =1;
                    /* burn front is stored in matrices nmat* */
                    /* and nnew* */
                    nmatx[ncur]=i;
                    nmaty[ncur]=j;
                    nmatz[ncur]=1;
                    /* Burn as long as new (fuel) pixels are found */
                    do{
                         nnew=0;
                         for(inew=1;inew< =ncur;inew+ +){
                              xcn=nmatx[inew];
                              ycn=nmaty[inew];
                              zcn=nmatz[inew];

                              /* Check all six neighbors */
                              for(jnew=1;jnew< =6;jnew+ +){
                                   x1=xcn;
                                   y1=ycn;
                                   z1=zcn;
                                   if(jnew= =1){x1-=1;}
                                   if(jnew= =2){x1+ =1;}
                                   if(jnew= =3){y1-=1;}
                                   if(jnew= =4){y1+ =1;}
                                   if(jnew= =5){z1-=1;}
                                   if(jnew= =6){z1+ =1;}

/* Nonperiodic so be sure to remain in the 3-D box */

if((x1> =1)&&(x1< =SYSSIZE)&&(y1> =1)&&(y1< =SYSSIZE)&&(z1> =1)&&
     (z1< = SYSSIZE)){
                                   if(cement [x1] [y1] [z1]= =npix){
                                    ntot+ =1;
                                    cement [x1] [y1] [z1]=BURNT;
                                        nnew+ =1;
                                        if(nnew> =9000){
                                         printf("error in size of nnew \n");
                                         }
                                     nnewx[nnew]=x1;
                                     nnewy[nnew]=y1;
                                     nnewz[nnew]=z1;
                         /* See if bottom of system has been reached */
```

```
                                        if(z1 = =SYSSIZE){igood = 1;}
                                    }
                                }
                                }
                            }
                            if(nnew>0){
                                ncur=nnew;
                                /* update the burn front matrices */
                                for(icur=1;icur< =ncur;icur+ +){
                                    nmatx[icur]=nnewx[icur];
                                    nmaty[icur]=nnewy[icur];
                                    nmatz[icur]=nnewz[icur];
                                }
                            }
                        }while (nnew>0);

                        ntop+ =ntot;
                        if(igood= =1){
                            nthrough+ =ntot;
                        }
                    }

            }
            }

            printf("Phase ID=  %d \n",npix);
            printf("Number accessible from top=  %ld \n",ntop);
            printf("Number contained in through pathways=  %ld \n",nthrough);

    /* return the burnt sites to their original phase values */
            for(i=1;i< =SYSSIZE;i+ +){
            for(j=1;j< =SYSSIZE;j+ +){
            for(k=1;k< =SYSSIZE;k+ +){

                if(cement [i] [j] [k]= =BURNT){
                    cement [i] [j] [k]=npix;
                }
            }
            }
            }
    }

    /* routine to assess the connectivity (percolation) of total solids */
    /* excluding aggregate when one is present in the system */
```

```
void consold()
{
        long int ntop,nthrough,ncur,nnew,ntot;
        int i,inew,j,k,nmatx[9000],nmaty[9000],nmatz[9000];
        int xcn,ycn,zcn,x1,y1,z1,igood,nnewx[9000],nnewy[9000],nnewz[9000];
        int jnew,icur;

/* counters for number of pixels of solids accessible from top surface */
/* and number which are part of a percolated pathway */
        ntop=0;
        nthrough=0;

        /* percolation is assessed from top to bottom only */
        /* and burning algorithm is nonperiodic in x and y directions */
        k=1;

        for(i=1;i<=SYSSIZE;i++){
        for(j=1;j<=SYSSIZE;j++){

                ncur=0;
                ntot=0;
                igood=0;    /* Indicates if bottom has been reached */
                if((cement [i] [j] [k]!=POROSITY)&&(cement [i] [j] [k]<12)&&(cement [i]
                   [j] [k]!=AGG)){
                /* burnt site has value 12 greater than original phase ID */
                        cement [i] [j] [k]+=12;
                        ntot+=1;
                        ncur+=1;
                        /* burn front is stored in matrices nmat* */
                        /* and nnew* */
                        nmatx[ncur]=i;
                        nmaty[ncur]=j;
                        nmatz[ncur]=1;

                        /* Burn as long as new (fuel) pixels are found */
                        do{
                                nnew=0;
                                for(inew=1;inew<=ncur;inew++){
                                        xcn=nmatx[inew];
                                        ycn=nmaty[inew];
                                        zcn=nmatz[inew];

                                        /* Check all six neighbors */
                                        for(jnew=1;jnew<=6;jnew++){
```

```
                                        x1=xcn;
                                        y1=ycn;
                                        z1=zcn;
                                        if(jnew==1){x1-=1;}
                                        if(jnew==2){x1+=1;}
                                        if(jnew==3){y1-=1;}
                                        if(jnew==4){y1+=1;}
                                        if(jnew==5){z1-=1;}
                                        if(jnew==6){z1+=1;}

/* Nonperiodic so be sure to remain in original 3-D box */

if((x1>=1)&&(x1<=SYSSIZE)&&(y1>=1)&&(y1<=SYSSIZE)&&(z1>=1)&&
     (z1<=SYSSIZE)){
          if((cement [x1] [y1] [z1]!=POROSITY)&&(cement [x1] [y1] [z1]<12)
                &&(cement [x1] [y1] [z1]!=AGG)){
                                        ntot+=1;
                                        cement [x1] [y1] [z1]+=12;
                                            nnew+=1;
                                            if(nnew>=9000){
                                             printf("error in size of nnew \n");
                                             }
                                         nnewx[nnew]=x1;
                                         nnewy[nnew]=y1;
                                         nnewz[nnew]=z1;
                                  /* See if bottom has been reached */
                                         if(z1==SYSSIZE){igood=1;}
                                       }
                                  }
                                  }
                            }
                            if(nnew>0){
                                  ncur=nnew;
                                  for(icur=1;icur<=ncur;icur++){
                                       nmatx[icur]=nnewx[icur];
                                       nmaty[icur]=nnewy[icur];
                                       nmatz[icur]=nnewz[icur];
                                  }
                            }
                      }while (nnew>0);

                      ntop+=ntot;
                      if(igood==1){
                            nthrough+=ntot;
```

```
                        }
                }

        }
        }

        printf("For total solids \n");
        printf("Number accessible from top=  %ld \n",ntop);
        printf("Number contained in through pathways=  %ld \n",nthrough);

/* return the burnt sites to their original phase values */
        for(i=1;i<=SYSSIZE;i++){
        for(j=1;j<=SYSSIZE;j++){
        for(k=1;k<=SYSSIZE;k++){

                if(cement [i] [j] [k]>12){
                        cement [i] [j] [k]-=12;
                }
        }
        }
        }
}

main(){
        int userc;      /* User choice from menu */
        int nseed,ig,jg,kg;

        /* Display an introduction to the user */
        intro();

        printf("Enter random number seed value \n");
        scanf("%d",&nseed);
        printf("%d \n",nseed);
        seed=(&nseed);

/* Initialize counters and system parameters */
        nfill=0;
        ncement=0;
        ncemdis=0;
        npr=0;
        aggsize=0;

/* clear the 3-D system to all porosity to start */
        for(ig=1;ig<=SYSSIZE;ig++){
```

```
        for(jg=1;jg<=SYSSIZE;jg++){
        for(kg=1;kg<=SYSSIZE;kg++){
                cement [ig] [jg] [kg]=POROSITY;
        }
        }
        }

/* present menu and execute user choice */
        do{
                printf(" \n Input User Choice \n");
                printf("1) Add a flat inert aggregate to microstructure \n");
                printf("2) Add spherical particles (C3S or filler) to microstructure \n");
                printf("3) Add one-pixel filler particles to microstructure \n");
                printf("4) Hydrate microstructure \n");
                printf("5) Measure phase fractions \n");
                printf("6) Measure phase fractions as a function \n");
                printf("   of distance from aggregate surface \n");
                printf("7) Measure single phase connectivity \n");
                printf("8) Measure total solids connnectivity \n");
                printf("9) Exit \n");

                scanf("%d",&userc);
                printf("%d \n",userc);

                switch (userc) {

                                case 1:
                                        addagg();
                                        break;
                                case 2:
                                        create();
                                        break;
                                case 3:
                                        filler();
                                        break;
                                case 4:
                                        hydrate();
                                        break;
                                case 5:
                                        measure();
                                        break;
                                case 6:
                                        measagg();
                                        break;
```

```
                    case 7:
                            connect();
                            break;
                    case 8:
                            consold();
                            break;
                    default:
                            break;
                    }
            } while (userc<9);
}
```

APPENDIX D
Modifying HYDRA3D

It is recognized that other cement researchers may want to use HYDRA3D as a starting point for developing their own simulations of material microstructure and properties. This appendix will attempt to provide a few guidelines for this process.

1) Maintain an original version of the code.

The user should not modify the original version of HYDRA3D, but rather should copy the source file to a new file and modify the new file to suit their needs. In this manner, the original code is always available as a baseline.

2) Limit the scope of modifications.

Since HYDRA3D has been developed in a modular fashion, the user should be able to make changes without changing every module in the program. For example, to change the shape of particles (from spheres to ellipses for instance), the user should only need to change the routines CREATE, GSPHERE, and CHKSPH to create the appropriately shaped particles. Since all routines operate at the pixel level, the hydration and analysis routines are independent of initial particle shape. To change the characteristics of the mineral admixture particles, the user would modify the routines MVCHANT and possibly ADDEXT to reflect the new reaction and volume stoichiometry of the pozzolanic reaction.

New analysis routines can simply be added as new modules, with a new menu selection added to allow the user to select the new feature. The three-dimensional microstructural representation is available globally so that any new module can easily access individual pixels of the current microstructure.

3) Make changes in an incremental fashion

If a user intends to make several modifications to the code, they should be made sequentially as opposed to concurrently. Although interactions between modifications are always a concern, sequential modification should limit the time needed to develop and debug the new (changed) program.

4) Understand the existing code before modifying it

Above all, the user should be sure that they understand the workings of the existing code before attempting any modifications!

APPENDIX E
System Requirements

The system configuration needed to successfully execute the HYDRA3D program will depend largely on the computing environment. Since the program is designed to implement a rather large scale simulation (one million pixel elements), the system requirements are not trivial. Sample system configurations at NIST on which HYDRA3D has been successfully implemented are outlined in Table II. While the source code file size remains constant from system to system, the executable code file size and memory needed for execution are both functions of the system being considered, as the default implementation of integer and real variable sizes will vary from computer to computer. At a minimum, it appears that 4 Mb of system memory would be required to execute HYDRA3D without extensive paging. Disk space requirements are generally minimal.

Table II

Memory Requirements for HYDRA3D

Computer System	Language	Source Code File Size (kilobytes)	Executable File Size (kilobytes)	Memory for Execution (Megabytes)
CONVEX C120	Fortran	38	264	11
CONVEX C120	C	30	74	3.5
SUN 3/160 ffpa accel.	C	30	41	3.3
CRAY Y-MP	Fortran	38	833	21.5
CRAY Y-MP	C	30	13228	14.2

The time required to execute HYDRA3D will naturally depend on the "problem" being simulated. However, to provide some idea of time requirements, Table III lists the execution times for the Fortran and C versions of HYDRA3D on a variety of computers. All times are those necessary to execute example 2 (w/c=0.5) from Section 4.2 of this report. The times may seem quite large, but it should be kept in mind that HYDRA3D simulates the complex process of cement hydration for a relatively large system, not a trivial task.

Table III

Timing Benchmarks for HYDRA3D
(Example 2 from Section 4.2)

Computer System	Programming Language	Execution Time (seconds)
SUN 3/160 (ffpa)	C	35932
CONVEX C120	C	9159
CONVEX C120	Fortran	9009
CRAY Y-MP	C	2197
CRAY Y-MP	Fortran	1123

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11. ABSTRACT (A 200-WORD OR LESS FACTUAL SUMMARY OF MOST SIGNIFICANT INFORMATION. IF DOCUMENT INCLUDES A SIGNIFICANT BIBLIOGRAPHY OR LITERATURE SURVEY, MENTION IT HERE.)

A computer program, HYDRA3D, to simulate cement microstructural development and quantify microstructural characteristics has been developed. HYDRA3D is a menu drive program available in either Fortran or C which allows a user to create a starting microstructure, execute hydration, measure phase fractions, and assess phase connectivity. This manual outlines the conceptual model on which HYDRA3D is based, describes the programs in detail, and provides examples of program usage. A system calling diagram, source code listings, guidelines for modifying the programs, and system requirements are provided in the Appendices.

12. KEY WORDS (6 TO 12 ENTRIES; ALPHABETICAL ORDER; CAPITALIZE ONLY PROPER NAMES; AND SEPARATE KEY WORDS BY SEMICOLONS)

Cement; computer modelling; hydration; interfacial zone; microstructure; percolation; simulation; software

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