A Micromechanics-Based Model for Stiffness and Strength Estimation of Cocciopesto Mortars

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Abstract

The purpose of this paper is to propose an inexpensive micromechanics-based scheme for stiffness homogenization and strength estimation of mortars containing crushed bricks, known as cocciopesto. The model utilizes the Mori-Tanaka method to determine the effective stiffness, combined with estimates of quadratic invariants of the deviatoric stresses inside phases to predict the compressive strength. Special attention is paid to the representation of C-S-H gel layer around bricks and interfacial transition zone around sand aggregates, which renders the predictions sensitive to particle sizes. Several parametric studies are performed to demonstrate that the method correctly reproduces data and trends reported in available literature. Moreover, the model is based exclusively on parameters with clear physical or geometrical meaning and as such it provides a convenient framework for its further experimental validation.

Keywords. micromechanics, homogenization, strength estimation, cocciopesto, C-S-H gel coating, iterfacial transition zone

1 Introduction

The use of lime as a binder in mortars is associated with well-known inconveniences such as slow setting and carbonation, high drying shrinkage and porosity and low mechanical strength [1]. Although these limitations have been overcome with the use of Portland cement in the last 50 years, lime mortars still find use in the restoration of historic structures. This is mainly due to their superior compatibility with the original materials in contrast to many modern renovation render systems, e.g. [14, 15, 27].

Mechanical properties of lime mortars can be improved by a suitable design of the mixture. Phoenicians were probably the first ones who added crushed clay products, such as burnt bricks, tiles or pieces of pottery, to lime mortars to increase their durability and strength. Romans called such material cocciopesto and utilized this mortar in areas where other natural pozzolans were not available. The cocciopesto-based structures exhibit increased ductility, leading to their remarkable resistance to earthquakes [3, 19].

Much later, it was found that the mortars containing crushed clay bricks, burnt at 600–900°C, exhibit a hydraulic character, manifested by the formation of a thin layer of Calcium-Silicate-Hydrate (C-S-H) gel at the lime-brick interface [18]. Since C-S-H gel is the key component responsible for favorable mechanical performance of Portland cement pastes [21], it is generally conjectured that the enhanced performance of cocciopesto mortars can be attributed to the high strength and stiffness of the C-S-H gel coating [3, 18, 19, 27]. This mechanism competes with
the formation of the Interfacial Transition Zone (ITZ) at the matrix-aggregate interface, known to possess higher porosity and thus lower stiffness in cement-based mortars, e.g. [22, 26, 34].

The purpose of this work is to interpret these experimental findings by a micromechanical model based on the Mori-Tanaka method [17], motivated by its recent applications to related material systems. These include, for example, estimates of effective thermal conductivity of rubber-reinforced cement composites [29], elasticity predictions for early-age cement [5] or alkali-activated [33] pastes, upscaling the compressive strength of cement mortars [24], and multi-scale simulations of three-point bending tests of concrete specimens [32]. Here, we exploit these developments to propose a simple analytical model for stiffness and strength estimation of cocciopesto mortars in Section 2. In particular, the elasticity predictions utilize Benveniste’s reformulation [4] of the Mori-Tanaka method [17], whereas the strength predictions build on recent results by Pichler and Hellmich [24], who demonstrated that compressive strength is closely related to the quadratic average of the deviatoric stress in the weakest phase. A particular attention is paid to representation of coatings by C-S-H gel and ITZ, which renders the predictions sensitive to the size of brick particles and aggregates. In Section 3, we verify predictions of the proposed scheme against data available in open literature. These finding are summarized in Section 4, mainly as a support for future validation of the model against experimental results. Finally, in Appendix A we gather technical details needed to account for coated inclusions in order to make the paper self-contained.

In what follows, the Mandel representation of symmetric tensorial quantities is systematically employed, e.g. [16, p. 23]. In particular, italic letters, e.g. \( a \) or \( A \), refer to scalar quantities and boldface letters, e.g. \( \mathbf{a} \) or \( \mathbf{A} \), denote vectors or matrix representations of second- or fourth-order tensors. \( \mathbf{A}^T \) and \( (\mathbf{A})^{-1} \) standardly denote the matrix transpose and the inverse matrix. Other symbols and abbreviations are introduced in the text when needed.

2 Model

We consider a composite sample occupying domain \( \Omega \), composed of \( n \) distinct phases indexed by \( r \). The value \( r = 0 \) is reserved for the matrix phase and \( r = 1, \ldots, n \) refer to heterogeneities having the shape of a sphere or spherical shell, see Fig. 1. Volume fraction of the \( r \)-th phase are defined as \( c^{(r)} = |\Omega^{(r)}|/|\Omega| \), where \( |\Omega^{(r)}| \) denotes the volume occupied by the \( r \)-th phase, and geometry of coated particles is specified by their radii \( R^{(r)} \) for \( r = 2, \ldots, 5 \), Fig. 2.

Elastic properties of individual phases are specified by the material stiffness matrix \( \mathbf{L}^{(r)} \). As each phase is assumed to be homogeneous and isotropic, we have

\[
\mathbf{L}^{(r)} = 3K^{(r)} \mathbf{I}_V + 2G^{(r)} \mathbf{I}_D \quad \text{for } r = 0, \ldots, n,
\]

where \( K^{(r)} \) and \( G^{(r)} \) are the bulk and shear moduli of the \( r \)-th phase, and \( \mathbf{I}_V \) and \( \mathbf{I}_D \) denote...
the orthogonal projections to the volumetric and deviatoric components, e.g. [12], so that
\[
\varepsilon(x) = (I_V + I_D)\varepsilon(x) = \varepsilon_V(x) + \varepsilon_D(x),
\]
\[
\sigma(x) = (I_V + I_D)\sigma(x) = \sigma_V(x) + \sigma_D(x),
\] 
(2a) (2b)
for \( x \in \Omega \). In Eq. (2), \( \varepsilon \) and \( \sigma \) refer to local stresses and strains, \( \varepsilon_V \) and \( \varepsilon_D \) are the volumetric and deviatoric strain components, \( \sigma_V \) and \( \sigma_D \) refer to the stress components and \( I \) is the second-order unit tensor (in the matrix representation).

Development of the model follows the standard routine of the continuum micromechanics, e.g. [35]. The sample \( \Omega \) is subjected to the overall strain loading \( E \). Neglecting the interaction among phases, the mean strains inside heterogeneities are obtained as
\[
E^{(r)} = A^{(r)} \varepsilon \quad \text{for} \quad r = 1, \ldots, n,
\]
where \( A^{(r)} \) is the dilute concentration factor of the \( r \)-th phase, see Section 2.1. In Section 2.2, after accounting for the phase interaction, these are combined to the full concentration factors satisfying
\[
E^{(r)} = A^{(r)} E \quad \text{for} \quad r = 0, \ldots, n,
\]
utilized next to estimate the overall stiffness of the composite material, \( L_{\text{eff}} \). Moreover, as outlined in Section 2.3, expression for the overall stiffness also encodes the mean value of the quadratic invariant of the local stress deviator \( \sigma_D \) defined as
\[
J_2^{(r)} = \sqrt{\frac{1}{2|\Omega^{(r)}|} \int_{\Omega^{(r)}} \sigma_D(x)^T \sigma_D(x) \, dx},
\] 
(3)
that can be directly used to estimate the overall strength of a material.

2.1 Dilute concentration factors

Due to geometrical and material isotropy of individual phases, the dilute concentration factors attain the form analogous to (1):
\[
A^{(r)}_{\text{dil}} = A^{(r)}_{\text{dil}, V} I_V + A^{(r)}_{\text{dil}, D} I_D \quad \text{for} \quad r = 1, \ldots, n.
\] 
(4)
The expressions for the components are given separately for the uncoated \( (r = 1) \) and coated \( (r = 2, \ldots, 5) \) particles. Namely, in the first case it holds
\[
A^{(1)}_{\text{dil}, V} = \frac{K^{(0)}}{K^{(0)} + \alpha^{(0)}(K^{(1)} - K^{(0)})},
\]
\[
A^{(1)}_{\text{dil}, D} = \frac{G^{(0)}}{G^{(0)} + \beta^{(0)}(G^{(1)} - G^{(0)})},
\]
with the auxiliary factors following from the Eshelby solution [9] in the form
\[
\alpha^{(0)} = \frac{1 + \nu^{(0)}}{3(1 + \nu^{(0)})},
\]
\[
\beta^{(0)} = \frac{2(4 - 5\nu^{(0)})}{15(1 - \nu^{(0)})},
\]
where \( \nu^{(0)} \) is the Poisson ratio of the matrix phase.

The coated case is more involved, and was first solved in its full generality by Herve and Zaoui [10] for multi-layered spherical inclusion. To apply their results in the current setting, we locally number the phases by the index \( i = [i_1, i_2, i_3]^T \), see Fig. 2, where \( i = [2, 3, 0]^T \) for the
Table 1: Reference properties of individual phases; \(\rho\) denotes density, \(f_t\) is tensile strength, \(m\) is the mass fraction.

| \(r\) | Phase | \(\rho\) [kgm\(^{-3}\)] | \(E\) [MPa] | \(\nu\) | \(f_t\) [MPa] | \(m\) | \(R\) [\(\mu\)m] | Note |
|-------|-------|----------------|---------|-------|-------------|------|----------|------|
| 0     | Pure lime matrix | 1,200\(^a\) | 2,000\(^b\) | 0.25\(^b\) | 0.4\(^b\) | 3 | \(\times\) | \(c^{(1)} = 35\%^a\) |
| 1     | Voids | \(\times\) | \(10^{-9}\) | 0.25 | \(\times\) | \(\times\) | | |
| 2     | Clay brick | 2,300\(^a\) | 5,000\(^a\) | 0.17\(^a\) | 3.2\(^a\) | 1 | 500 | |
| 3     | C-S-H gel | 2,000\(^[11]\) | 22,000\(^[7]\) | 0.2\(^[7]\) | \(\times\) | \(\times\) | 510\(^[6]\) | |
| 4     | Siliceous sand | 2,700\(^a\) | 60,000\(^{[32]}\) | 0.17\(^{[32]}\) | 48\(^{[21]}\) | 1 | 500 | |
| 5     | ITZ | 1,200\(^b\) | 500\(^c\) | 0.25\(^b\) | \(\times\) | \(\times\) | 520\(^c\) | |

\(^a\) Our own (unpublished) data.
\(^b\) Same value as for the lime matrix.
\(^c\) Set as in \([34]\) for cement-based concretes, i.e., Young’s modulus to 20–40% of the value for the matrix phase and thickness to 20 \(\mu\)m.

brick–C-S-H conglomerate and \(i = [4, 5, 0]^T\) refers to a sand particle coated by ITZ. Now, we have

\[
A^{(i_1)}_{\text{dil}, V} = \frac{1}{Q_{11}^2}, \quad A^{(i_2)}_{\text{dil}, V} = \frac{Q_{11}^1}{Q_{11}^2},
\]

and

\[
A^{(i_1)}_{\text{dil}, D} = A_1 - \frac{21}{5} \frac{R^{(i_1)2} - R^{(i_1)} B_1}{1 - 2\nu^{(i_1)}},
\]

\[
A^{(i_2)}_{\text{dil}, D} = A_2 - \frac{21}{5} \frac{R^{(i_2)5} - R^{(i_2-1)5}}{(1 - 2\nu^{(i_2)})(R^{(i_2)3} - R^{(i_2-1)3})} B_2,
\]

where the auxiliary factors are provided in Appendix A.

### 2.2 Stiffness estimates

In Benveniste’s \([4]\) interpretation of the original Mori-Tanaka method \([17]\), the mutual interaction among heterogeneities is modeled by loading each particle by the average strain in the matrix phase \(E^{(0)}\) instead of \(E\). For this purpose, we relate \(E^{(0)}\) to \(E\) by a strain compatibility condition, valid under the dilute approximation,

\[
E = \left( c^{(0)} I + \sum_{r=1}^{n} c^{(r)} A^{(r)}_{\text{dil}} \right) E^{(0)},
\]
from which we express the full concentration factors as

\[ A^{(0)} = (c^{(0)} I + \sum_{r=1}^{n} c^{(r)} A^{(r)}_{\text{dil}})^{-1}, \]

\[ A^{(r)} = A^{(r)}_{\text{dil}} A^{(0)} \text{ for } r = 1, \ldots, n. \]

Utilizing a universal relation

\[ L_{\text{eff}} = \sum_{r=0}^{n} c^{(r)} L^{(r)} A^{(r)}, \]

we can see that the effective stiffness inherits the symmetry of individual phases (1) with

\[ K_{\text{eff}} = \frac{c^{(0)} K^{(0)} + \sum_{r=1}^{n} c^{(r)} K^{(r)} A^{(r)}_{\text{dil},V}}{c^{(0)} + \sum_{r=1}^{n} c^{(r)} A^{(r)}_{\text{dil},V}}, \tag{7a} \]

\[ G_{\text{eff}} = \frac{c^{(0)} G^{(0)} + \sum_{r=1}^{n} c^{(r)} G^{(r)} A^{(r)}_{\text{dil},D}}{c^{(0)} + \sum_{r=1}^{n} c^{(r)} A^{(r)}_{\text{dil},D}}. \tag{7b} \]

### 2.3 Strength estimates

As recognized first by Kreher [13], the fluctuations of stresses and strains in individual phases can be estimated from the energy conservation condition due

\[ E^T L_{\text{eff}} E = \frac{1}{|\Omega|} \sum_{r=0}^{n} \left( 9 K^{(r)} \int_{\Omega^{(r)}} \varepsilon_{V}^2(x) \, dx + \int_{\Omega^{(r)}} 2 G^{(r)} \varepsilon_{D}^T(x) \varepsilon_{D}(x) \, dx \right), \tag{8} \]

expressing the conservation of energy on macroscale due to \( E \) and the average local values due to \( \varepsilon_{V} \) and \( \varepsilon_{D} \). Differentiating (8) with respect to \( G^{(r)} \), we obtain

\[ E^T \frac{\partial L_{\text{eff}}}{\partial G^{(r)}} E = 2 \frac{1}{|\Omega|} \int_{\Omega^{(r)}} \varepsilon_{D}^T(x) \varepsilon_{D}(x) \, dx, \]

for \( r = 0, \ldots, n \). Next, we recognize that \( \sigma_{D}^{(r)} = (1/2 G^{(r)}) \varepsilon_{D}^{(r)} \) and recall the definition of the quadratic invariant (3) to arrive at

\[ J_2^{(r)} = G^{(r)} \sqrt{\frac{1}{c^{(r)}} E^T \frac{\partial L_{\text{eff}}}{\partial G^{(r)}} E}. \tag{9} \]

As thoroughly demonstrated by and Pichler and Hellmich [24], this quantity is closely related to the compressive strength \( f_c \) of cement pastes at various degrees of hydration. Here, we postulate that

\[ \frac{f_c(p_1)}{f_c(p_2)} \approx \frac{J_2^{(w)}(p_2)}{J_2^{(w)}(p_1)}, \tag{10} \]

where \( w = 0, \ldots, n \) is the index of the weakest phase and \( p \) refers to a parameter characterizing the mixture composition, see the next section for concrete examples.
3 Results and discussion

The purpose of this section is to examine the trends in mechanical properties as predicted by the proposed scheme. Default data for individual phases, summarized in Table 1, were partly assembled from open literature and complemented with our own, yet unpublished, measurements. Note that the matrix–brick–sand fractions correspond to a typical composition of historic lime mortars [3, 2] and that the engineering constants $E$ and $\nu$ are connected to the bulk and shear moduli through well-known relations, e.g. [16, p. 23],

$$K = \frac{E}{3(1 - 2\nu)}, \quad G = \frac{E}{2(1 + \nu)}.$$  

, motivated by experimental findings in e.g. [3, 31, 30], we assume that an increase in the C-S-H gel volume fraction ($\Delta c^{(3)}$) is compensated by the corresponding changes for matrix ($\Delta c^{(0)}$), voids ($\Delta c^{(1)}$), and clay bricks ($\Delta c^{(2)}$), so that

$$\Delta c^{(0)} + \Delta c^{(1)} + \Delta c^{(2)} + \Delta c^{(3)} = 0,$$

where we set for simplicity $\Delta c^{(1)} = \Delta c^{(2)} = \Delta c^{(3)}$. Analogously, an increase in the volume fraction of ITZ corresponds to the decrease in volume of the matrix phase:

$$\Delta c^{(0)} + \Delta c^{(5)} = 0.$$

In the strength estimates, the imposed loading simulates the uniaxial compression test, for which $\Sigma = [−1, 0, 0, 0, 0]^T$ and the average strain follows from

$$E = (L_{\text{eff}})^{-1} \Sigma.$$  

We assume that ITZ is the weakest phase, i.e. $w = 5$ in Eq. (10), and estimate the derivative in Eq. (9) by the forward difference with the step size of $\Delta G^{(5)} = 1 \text{ Pa}$.³

3.1 Effect of coatings

The first aspect we would like to discuss is the effect of coating on the dilute concentration factors of the brick and sand particles. Fig. 3 demonstrates that, in terms of the volumetric phase strains, the effects of C-S-H and ITZ are comparable, despite the fact the C-S-H is stiffer and ITZ more compliant than the matrix phase. The differences in the deviatoric part, which drive the strength estimates according to Eq. (10), become more pronounced with the increasing thickness. This indicates that the contribution of brick and sand particles to the overall properties might still be different, once accounting phase properties and their interaction, see Section 3.3 for further discussion.

3.2 Influence of porosity

Analogously to the cement pastes, porosity has a major influence on the overall properties of lime-based mortars. This is also confirmed by the results of the proposed model shown in Fig. 4. As for the overall stiffness, for the realistic range of porosities of 25–40% [8], the estimates (7) predict the values of Young modulus between $\approx 2,000$ and $1,000 \text{ MPa}$. This is consistent with the values reported in [3] historic lime mortars (without pozzolan admixtures).

As for the strength estimates, it follows from Fig. 4 (b) that they reproduce [21, p. 280]

$$f_c(c^{(1)}) = f_c(0)(1 - c^{(1)})^n,$$  

(11)

³ Our results are reproducible with a MATLAB code Homogenizator MT, freely available at http://mech.fsv.cvut.cz/~nezerka/software.
with $n = 1.04$, yielding practically the linear strength-porosity scaling. Unfortunately, we are currently unable to validate this prediction against experiments; the only available work we are aware of by Papayianni and Stefanidou [23] does not contain enough data. Still, Eq. (11) complies with the fact the influence of porosity is much smaller in lime mortars than in cement-based materials, for which $n \approx 3$ is typically used.

### 3.3 Size effects

Now we proceed to clarify the impact of brick and sand particles on the overall mechanical properties. In particular, when increasing the size of brick particles, the material becomes more compliant since the stiffening effect of C-S-H layer decreases, Fig. 5(a). This also increases the deviatoric stresses in ITZ, as manifested by the strength reduction visible in Fig. 5(b). These effects practically stabilize for particles larger than 0.5 mm and their magnitude is rather limited: the stiffness decreases by about 10% and the strength only by 4%. Such trends are qualitatively consistent with the results presented e.g. in [3, 19, 27].

Larger sand particles, on the other hand, tend to make the composite material stiffer, Fig. 6(a), by compensating for inferior mechanical properties of ITZ. Since the relative thickness of ITZ layer decreases, the stresses inside this phase increase and the material becomes weaker in overall, Fig. 6(b). When compared to brick particles, these effects are much more pronounced: in the considered range of radii, the Young modulus increases by about 100% and the strength decreases by 25% with no tendency to stabilize. This agrees well with experimental outcomes reported in [28].

### 4 Conclusions

In the present work, following the recent developments presented in [25, 24, 33, 32], a simple micromechanics-based scheme for strength and stiffness estimates of cocciopesto mortars has
been presented. The model utilizes directly measurable material and geometrical properties of individual phases and is free of adjustable parameters. On the basis of presented results, we conclude that the model

1. predicts realistic values of the overall Young modulus and the strength-porosity scaling,
2. captures the “smaller is stiffer” and “smaller is stronger” trends for crushed brick particles,
3. captures the “larger is stiffer” and “larger is weaker” trends for sand aggregates,
4. explains the positive role of crushed bricks in comparison with sand aggregates.

Of course, in order to accept this model for practical use, it needs to be validated against comprehensive experimental data at micro- and macro-scales. This topic is currently under investigation and will be reported separately.

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A Herve-Zaouï solution

The effect of coating on the mechanical properties enters the solution through the auxiliary factors $Q^k$ in Eq. (5), and $A^k$ and $B^k$ in Eq. (6). Here, these are provided in the closed form optimized for coding, utilizing the results and nomenclature by Herve and Zaouï [10]. Note that in order to keep the notation consistent, $a^{(k)}$ corresponds to a property of the $k$-th phase, whereas $a^k$ denotes a quantity utilized in the Herve-Zaouï solution (independent of $a^{(k)}$). Also recall that we employ the local numbering of phases by the index $i = [i_1, i_2, i_3]^T$ introduced by Fig. 2.

In particular, the volumetric part is expressed in terms of matrices

$$Q^1 = N^1, \quad Q^2 = N^2 Q^1,$$

with

$$N^k = \frac{1}{3K^{(i_{k+1})} + G^{(i_{k+1})}} \begin{bmatrix} 3K^{(i_k)} + 4G^{(i_{k+1})} & 4 \frac{R^{(i_k)}3}{3K^{(i_{k+1})} + 4G^{(i_k)}} \end{bmatrix} \text{ for } k = 1, 2.$$  

The matrices needed to evaluate the deviatoric part follow from

$$A^1 = \frac{P_{22}^2}{P_{21}^2 P_{22}^2 - P_{21}^2 P_{21}^2}, \quad A^2 = W_1^2, \quad B^1 = \frac{-P_{21}^2}{P_{11}^2 P_{22}^2 - P_{12}^2 P_{21}^2}, \quad B^2 = W_2^2,$$

where

$$W^k = \frac{1}{P_{22}^2 P_{21}^2 - P_{21}^2 P_{22}^2} P^{k-1} \begin{bmatrix} P_{22}^2 & -P_{21}^2 & 0 \end{bmatrix}^T (k = 1, 2), \quad P^1 = M^1, \quad P^2 = M^2 P^1.$$  

The auxiliary matrix $M^k$ admits the expression:

$$M^k = \frac{1}{5(1 - \nu^2)} \begin{bmatrix} \frac{c^k}{3} & R^{(i_k)}2(6^k - 7^k) & -12\alpha^k & \frac{4(f^k - 27\alpha^k)}{15R^{(i_k)}3(1 - 2\nu^2)} \frac{M_{34}}{7R^{(i_k)}} & \frac{M_{23}}{7} \frac{M_{34}}{3(1 - 2\nu^2)} \\ \frac{3}{} & b^k(1 - \nu^2(i_k + 1)) & 5(1 - 2\nu^2(i_k)) & \frac{12\alpha^k}{15R^{(i_k)}3(1 - 2\nu^2)} & \frac{M_{23}}{7} \frac{M_{34}}{3(1 - 2\nu^2)} \\ 0 & \frac{R^{(i_k)}2}{70(1 - 2\nu^2)} & \frac{R^{(i_k)}2(2^k + 14\alpha^k)}{70(1 - 2\nu^2)} & \frac{M_{23}}{7} \frac{M_{34}}{3(1 - 2\nu^2)} \frac{M_{41}}{3} & \frac{M_{34}}{3(1 - 2\nu^2)} \\ \frac{10}{M_{41}} & \frac{7\alpha^k R^{(i_k)}2(1 - 2\nu^2)}{2(1 - 2\nu^2)} & \frac{10}{M_{41}} & \frac{M_{23}}{7} \frac{M_{34}}{3(1 - 2\nu^2)} \frac{M_{41}}{3} & \frac{M_{34}}{3(1 - 2\nu^2)} \\ \end{bmatrix}$$

with

$$M_{23} = \frac{-20\alpha^k(1 - 2\nu^2(i_k + 1))}{7R^{(i_k)}7}, \quad M_{41} = \frac{-5\alpha^k R^{(i_k)}3(1 - 2\nu^2(i_k + 1))}{6},$$

$$M_{34} = \frac{R^{(i_k)}2(105(1 - \nu^2(i_k + 1)) + 12\alpha^k(7 - 10\nu^2(i_k + 1)) - 7e^k)}{35(1 - 2\nu^2)}.$$
and

\[ a^k = \frac{G^{(i_k)}}{G^{(i_{k+1})}}(7 + 5G^{(i_k)})(7 - 10G^{(i_{k+1})}) - (7 - 10G^{(i_k)})(7 + 5G^{(i_{k+1})}), \]

\[ b^k = \frac{G^{(i_k)}}{G^{(i_{k+1})}}(7 + 5G^{(i_k)}) + 4(7 - 10G^{(i_k)}), \]

\[ c^k = (7 - 5G^{(i_{k+1})}) + 2(4 - 5G^{(i_{k+1})})\frac{G^{(i_k)}}{G^{(i_{k+1})}}, \]

\[ d^k = (7 + 5G^{(i_{k+1})}) + 4(7 - 10G^{(i_{k+1})})\frac{G^{(i_k)}}{G^{(i_{k+1})}}, \]

\[ e^k = 2(4 - 5G^{(i_k)}) + \frac{G^{(i_k)}}{G^{(i_{k+1})}}(7 - 5G^{(i_k)}), \]

\[ f^k = (4 - 5G^{(i_k)})(7 - 5G^{(i_{k+1})}) - \frac{G^{(i_k)}}{G^{(i_{k+1})}}(4 - 5G^{(i_{k+1})})(7 - 5G^{(i_k)}), \]

\[ \alpha^k = \frac{G^{(i_k)}}{G^{(i_{k+1})}} - 1. \]