Finite element simulation of duplex compaction processes

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INTRODUCTION

The use of duplex fuel pellets that consist of an inner core and outer ring has been considered for high burnup nuclear fuel.\(^1\text{-}\text{4}\) The use of different pairs of materials, such as UO\(_2\) and ThO\(_2\),\(^5\text{-}\text{7}\) or enriched and natural UO\(_2\),\(^8\text{-}\text{10}\) for the inner and outer parts of the duplex pellet has been proposed to reduce the radial temperature and burnup distribution, which lead to a decrease in fission gas release and an increase in pellet burnup. Several processes have been used to make duplex pellets and can be classified into a separate process because green compacts of metal oxide powders are not usually as strong as sintered components. During compaction, it is important to minimise any local density difference in a compact since volume shrinkage is a function of density during sintering. Large differences in volume shrinkage in a compact can cause cracks or a bad shape to be formed after sintering. Heterogeneity of density is caused by friction force due to interparticle movement and relative slip between the powder particles and the die wall. Also the die geometry and the sequence of punch movements result in heterogeneity of density in a compact of complex shape. Since the compaction procedure in the combined process is not simple, prediction of density distribution in the duplex compact is important for proper tooling design.

Recently, the finite element method has offered a computational tool for simulation and analysis of the powder compaction process.\(^5\text{-}\text{9}\) For finite element simulation, a mathematical model of the powder compact is required. Two types of models that are generally used are the soil mechanics models and the isotropic hardening models. The cap model is popular among the soil mechanics models. This is a multisurface elastoplastic model permitting the representation of densification, hardening, as well as interparticle friction.\(^10\) On the other hand, models of the isotropic hardening type adopt the single surface elastoplastic method based on the associative flow rule and isotropic hardening. This type of model was originally proposed by Kuhn and Downey,\(^11\) Shima and Oyane,\(^12\) Doraiavelu et al.\(^13\) and Kim et al.\(^14\) also proposed empirical models of this type. Lee and Kim\(^15\) modified the yield criterion suggested by Doraiavelu et al.\(^13\) This could incorporate one empirical parameter that can be estimated from the yield stress versus the initial relative density data. Using the yield criterion, Han et al.\(^16\) formulated an elastoplastic finite element code and analysed the deformation of sintered porous metals in simple upsetting, indenting,\(^16\) ring compression\(^17\) and hot forging.\(^18\) Also, Han et al. calculated forging limit curves of sintered porous metals.\(^19\) Park et al. modified the yield criterion suggested by Lee and Kim,\(^15\) so that the modified yield criterion could successfully incorporate empirical parameters that reflect the characteristics of copper powders of different particle shape.\(^9\) Park et al. applied the yield criterion to ceramic powders assuming no strain hardening of powder material and showed that the density–compaction load curve of ceramic powders can be well fitted by the yield criterion.\(^7\)

The yield criterion makes a nonlinear equation with an elastoplastic constitutive equation. Therefore numerical time integration of the constitutive equation is necessary for finite element calculation. An integration scheme is important for the accuracy and stability of the solution, especially for problems of finite strain where plastic strains are two to three orders of magnitude larger than elastic strains. Ortiz and Popov\(^21\) presented a systematic study of the accuracy and stability of the generalised trapezoidal and generalised midpoint rules. Their results show that, for strain increments which are several times larger than the size of the yield surface in strain space, the backward Euler
method leads to better accuracy, whereas for strain increments smaller than the yield strain, the trapezoidal rule provides optimal accuracy. Aravas investigated the backward Euler method for the time integration of the constitutive equation with pressure dependent yield criteria.

In this paper, the yield criterion proposed by Park et al. is briefly explained and the application of the backward Euler integration algorithm to the yield criterion is discussed. Three combined types of duplex compaction process are simulated by a finite element code implemented using the backward Euler method with the yield criterion. Density distributions in duplex compacts of each compaction process are investigated.

YIELD CRITERION OF CERAMIC POWDER COMPACT

A number of authors proposed yield criteria of porous material which can be generalised in the following form: \[ A f^2 + B f^3 = \eta Y_0^2 = Y_R^2 \]

where \[ \eta = Y_R^2 / Y_0^2, \quad J_1 = \sigma_{11} + \sigma_{22} + \sigma_{33} \]

\[ J_2 = \frac{1}{6} \left[ (\sigma_{11} - \sigma_{22})^2 + (\sigma_{22} - \sigma_{33})^2 + (\sigma_{33} - \sigma_{11})^2 \right] + \sigma_{12}^2 + \sigma_{13}^2 + \sigma_{23}^2 \]

\[ J_3, \quad J_1 \] and \( R \) are second deviatoric stress invariant, first stress invariant and relative density, respectively; \( A, B \) and \( \eta \) are functions of relative density; \( Y_0 \) is the yield stress of porous material having relative density \( R \) and \( Y_0 \) is the yield stress of the base material.

Expressions for \( A, B \) and \( \eta \) of the proposed yield criteria are summarised in Table 1.

Park et al. proposed an empirical yield criterion of metal oxide powder compact.

\[ (2 + R^2) J_2 + \frac{1}{3} R^2 J_3 = \frac{1}{R_T} \left( \frac{R - R_T}{1 - R_T} \right)^m K^2 = \eta Y_0^2 \]

where \( R_T \) and \( m \) are the relative tap density and geometric hardening exponent, respectively. They are experimental parameters varying with the characteristics of powder such as shape and size distribution. \( Y_0 \) is assumed to be constant since plastic deformation of metal oxide powder particles is negligible. The yield surface given by equation (2) becomes that of von Mises for relative density \( R = 1 \). When a powder compact has a relative density \( R \) which is less than unity, there is an effect of the hydrostatic pressure on the plastic flow as shown in Fig. 1. The yield criterion given by equation (2) would be a useful model only for densification processes such as hydrostatic pressing and uniaxial compaction.

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Expressions for ( A, B ) and ( \eta )</th>
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<tbody>
<tr>
<td>A(R)</td>
<td>B(R)</td>
</tr>
<tr>
<td>Shima and Oyane 12</td>
<td>3</td>
</tr>
<tr>
<td>Gurson 13</td>
<td>5+R</td>
</tr>
<tr>
<td>Doraisel et al 13</td>
<td>2+R^2</td>
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<tr>
<td>Lee and Kim 14</td>
<td>2+R^2</td>
</tr>
<tr>
<td>Park et al 15</td>
<td>2+R^2</td>
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</table>

1 Yield surface representation at various values of \( R \)

For uniaxial die compaction, the relation between the compaction pressure and relative density can be derived from equation (2). Assuming that there is no friction between powder compact and die wall, i.e. no shear stress, the stress state and strain rate of the powder compact during uniaxial die compaction become

\[ \sigma_{33} = P, \quad \sigma_{11} = \sigma_{22} = S \quad \text{and} \quad \sigma_{12} = \sigma_{23} = \sigma_{31} = 0 \]

\[ \epsilon_{11} = \epsilon_{22} = \epsilon_{33} = 0 \]

where \( P \) and \( S \) are uniaxial compaction stress and radial stress, respectively. The subscript \( 3 \) in equation (3) represents the axis parallel with a uniaxial compaction direction and the subscripts 1 and 2 represent the other two axes in the Cartesian coordinate system. Applying the associated flow rule to the yield criterion of equation (2), strain rate \( \dot{\epsilon} \) can be expressed as

\[ \dot{\epsilon} = \frac{\dot{R}}{2Y_R} \left[ (2 + R^2) \sigma_{ij} - R^2 \sigma_{kk} \delta_{ij} \right] \]

where

\[ \dot{R} = \frac{1}{2 + R^2} \left\{ \frac{2}{3} \left[ (\dot{\epsilon}_{11} - \dot{\epsilon}_{22})^2 + (\dot{\epsilon}_{22} - \dot{\epsilon}_{33})^2 + (\dot{\epsilon}_{33} - \dot{\epsilon}_{11})^2 \right] + \dot{\epsilon}_{12}^2 + \dot{\epsilon}_{13}^2 + \dot{\epsilon}_{23}^2 \right\} + \left( \frac{1}{3(1 - R^2)} \right) \left( \dot{\sigma}_{11} + 2\dot{\sigma}_{22} + 3\dot{\sigma}_{33} \right)^2 \]

Combining equations (3) and (4), the relation between \( S \) and \( P \) can be obtained.

\[ S = \frac{R^2}{2 - R^2} P \]

As the shear strain rate components are zero under frictionless conditions, the pressure applied to the top surface of the compact \( P \) can be given from equations (4) and (5).

\[ P = \left( \frac{R - R_T}{1 - R_T} \right)^m \left( \frac{(2 - R^2)}{1 - (2R^2)^m (2 + R^2)} \right)^{1/2} K \]

The effects of parameters \( m \) and \( K \) can be seen in Fig. 2. Figure 2a shows the variation of uniaxial compaction pressure \( P \) for different \( m \) values as a function of relative density when \( R_T = 0.3 \) and \( K = 4000 \) in equation (6). At high values of \( m \), the curve increases slowly at low relative density and increases rapidly at high relative density. At low values of \( m \), the curve increases more steadily than the curve of high \( m \) cases. Therefore, \( m \) characterises the shape of the pressure–relative density curve of uniaxial compaction. Figure 2b shows the variation of pressure for different \( K \) values as a function of relative density when \( R_T = 0.3 \) and \( m = 4 \) in equation (6). As \( K \) increases, pressure increases proportionally. Therefore, \( K \) characterises the level of uniaxial compaction pressure. The parameters \( m \) and \( K \)
can be determined for a specific powder and a specific tap density by fitting equation (6) to the uniaxial compaction data. The total strain \( \varepsilon \) is known at the end of the increment as a displacement field given by the virtual work principle. The elasticity equation gives

\[
\sigma_i = C_{ij} : \varepsilon^e = C_{ij} : \Delta \varepsilon^\rho \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots 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Projecting the elasticity equation (10) onto \( I \) and \( n_x \), and using equation (17), \( p \) and \( q \) can be given as

\[
p = p^0 + B_0 \Delta \epsilon_p \quad \text{and} \quad q = q^0 - 3G_0 \Delta \epsilon_q
\]  

(21)

Evolution of the relative density is related to the change of the total volume as

\[
\Delta R = -R_e \Delta \epsilon^P : I = -R_e \Delta \epsilon_p
\]  

(22)

In summary, the problem of integrating the elastoplastic constitutive equation requires the solution of the following set of nonlinear equations for \( \Delta \epsilon_p, \Delta \epsilon_q, p, q \) and \( \Delta R \):

\[
\Delta \epsilon_p \left( \frac{\partial \Phi}{\partial \epsilon} \right)_\epsilon + \Delta \epsilon_q \left( \frac{\partial \Phi}{\partial q} \right)_\epsilon = 0
\]  

(23)

\[
\Phi(p, q, R_e) = \frac{2 + R_e^2}{3} q^2 + 3(1 - R_e^2)p^2 - \left( \frac{R_e - R_T}{1 - R_T} \right)^m K^2 = 0
\]  

(24)

\[
p = p^0 + B_0 \Delta \epsilon_p
\]  

(25)

\[
q = q^0 - 3G_0 \Delta \epsilon_q
\]  

(26)

\[
\Delta R = -R_e \Delta \epsilon_p
\]  

(27)

These equations are solved using Newton’s iterative method. Selecting \( \Delta \epsilon_p \) and \( \Delta \epsilon_q \) as the primary unknowns, equations (23) and (24) become the basic equations, in which \( p, q \) and \( \Delta R \) are defined by equations (25)–(27). Using \( c_p \) and \( c_q \) as the corrections for \( \Delta \epsilon_p \) and \( \Delta \epsilon_q \), the Newton iterative method gives the following linear equations.

\[
A_{11} c_p + A_{12} c_q = b_1 \quad \text{and} \quad A_{21} c_p + A_{22} c_q = b_2
\]  

(28)

where the constants \( A_i \) and \( b_i \) are given in Appendix. These linear equations are solved for \( c_p \) and \( c_q \), and the values of \( \Delta \epsilon_p \) and \( \Delta \epsilon_q \) are then updated.

\[
\Delta \epsilon_q \rightarrow \Delta \epsilon_q + c_p \quad \text{and} \quad \Delta \epsilon_q \rightarrow \Delta \epsilon_q + c_q
\]  

(29)

The values of \( p \) and \( q \) are updated using equations (25) and (26). Finally, the increment of the relative density is updated by equation (27). This iterative loop is continued until \( \Delta \epsilon_q \) and \( \Delta \epsilon_q \) converge.

The numerical integration method for the constitutive relation and the yield criterion for ceramic powder compaction described in the above sections were implemented in the elastoplastic finite element code AMEC2D.24,25

### EXPERIMENTAL PROCEDURE

A commercial ZrO2 powder was used in this study. The relative tap density \( R_T \) of powder was measured by measuring volume and weight of the tapped powder. The measured relative tap density is 0.371. Single action uniaxial compaction experiments were carried out using a universal tension and compression testing system in the pressure range from 0 to 450 MPa. The diameter of the steel die was 15 mm and the crosshead speed was 2 mm min\(^{-1}\). After uniaxial compaction, the average green compact density was obtained by measuring dimensions and weight. During uniaxial die compaction, the compaction stroke and the load were measured and used for determination of parameters \( m \) and \( K \).

Three combined types of duplex compaction process were carried out using the ZrO2 powder. Figure 3 shows the type I duplex compaction process. At first, compaction of the outer ring is carried out until the average relative density achieves a final average relative density of 0.53. Then, compaction of the inner core in the upper ring punch is carried out until the same final average relative density is reached. Finally, pushing the inner core into the outer ring completes process I. The type II duplex compaction process is shown in Fig. 4. It differs from the type I process in that the inner and outer compacts are compacted when their relative density is lower than the final average relative density of 0.53. At first, the two parts undergo precompaction up to an average relative density of 0.45. After combination, the combined inner and outer compacts are compacted together, which is the cocompaction step. The duplex compaction process, type III, is similar to the type II process, but uses a different die geometry as shown in Fig. 5. The inner diameter of the upper ring punch is smaller than the diameter of the lower core punch by 0.16 mm. Consequently there exists a clearance between inner and outer precompacts during the combination step. The inner core diameter was 6.8 mm in the experiment.

Relative density distributions of a duplex compact produced by the type III process were measured using the Kuczynski and Zaplatynskyj method.26 In this method, the relationship between hardness and relative density is obtained from measuring hardness and relative densities of thin compacts produced under various compaction...
pressures. The density of the thin compacts is usually uniform throughout the compact. The ratio of height to diameter of thin compacts used in this experiment is below 0.2. Micro Vickers hardness was measured ten times on the sectioned surface and the average value was used. Figure 6 shows the relationship between hardness and relative density. An empirical expression of relative density as a function of hardness was obtained from linear fitting of the experimental data.

\[ R = 0.4 + 8.6 \times 10^{-4}H \]  \hspace{1cm} (30)

where \( H \) is Vickers hardness. The relative density distribution of a duplex compact was obtained by converting hardness at selected points of the sectioned compact to relative density using equation (30).

**SIMULATION OF DUPLEX COMPACTION PROCESSES**

In order to determine the material parameters, single action uniaxial die compaction was conducted with ZrO\(_2\) powder. Figure 7 shows the measured compaction pressure as a function of relative density during uniaxial compaction. Using a non-linear best fitting method with experimentally measured \( R_T \) of 0.371, the parameters \( m \) and \( K \) in equation (6) could be determined as 3.8 and 3820 MPa, respectively. An analytic solution and FEM solution using the backward Euler method for compaction pressure as a...
function of relative density were compared with the experimental data as shown in Fig. 7. The analytical and numerical results show good agreement with experimental results, indicating that the yield criterion can successfully describe the compaction behaviour of ZrO$_2$ powder. The finite element method was used for simulation of the three combined type duplex compaction processes that are described in previous section.

**Determination of friction coefficient between powder compacts**

The type I process was simulated by FEM using the initial mesh shown in Fig. 8. Half of the compaction system was modelled by axisymmetric elements. The elastic modulus and Poisson’s ratio used in calculation are 200 GPa and 0.23, respectively. As the ring and core separate during the combination process, it is necessary to determine the friction coefficient of contact interface between the inner and outer ZrO$_2$ compacts. Figure 9 compares the load measured at the inner core punch during the combination process with that calculated by FEM using four different values of friction coefficient: 0.1, 0.3, 0.5 and 0.7. During combination, the contact area between the two parts increases. Friction force $f$ is proportional to contact area, $A$ as

$$f = \mu SA$$

where $\mu$ and $S$ are the friction coefficient and the pressure normal to the contact surface, which is the radial stress at contact surface, respectively. Therefore, the combination load increases as contact area increases. The calculation data of the friction coefficient as 0.5 shows the best agreement with the experimental data. The friction coefficient of contact interface between inner and outer compacts was assumed to be 0.5 for all cases of FEM calculation.

**Effects of average relative density before combination (types I and II)**

Figure 10a shows calculated relative density distributions on right half of the duplex compacts for the type I process. In the type I process, the inner core and outer ring compacts are combined when their average relative density is 0.53. After combination, relative density is highest at the top of the inner core and lowest at the bottom of the inner core. The difference between minimum and maximum relative density is 0.061. In the outer ring, relative density shows a relatively even distribution compared with the inner core. The large difference of relative density in the inner core and the small difference of relative density in outer ring are caused by friction force that occurs at the contact surface. In general, powder compacts made by uniaxial die compaction have a density distribution that shows high density at the top and...
low density at the bottom owing to friction between the compact and the die wall. During combination of the inner core and the outer ring compacts, friction force occurs on the contact surfaces as shown in Fig. 11. The powder flow induced by the friction force goes upward in the inner core and goes downward in the outer ring. This opposite powder flow increases the relative density difference in the inner core and decreases that in the outer ring. It can also be seen that the height of the inner core becomes shorter in the axial direction after combination due to the friction force as denoted by an arrow in Fig. 10a. Figure 12 shows the top and bottom surfaces of a duplex compact, respectively and the height difference can be observed in Fig. 12b.

In order to reduce friction force, which causes height difference and gives rise to density difference during combination, it is necessary to reduce the pressure normal to the contact surface $S$ in equation (31), which is the radial stress at the contact surface. Radial stress decreases as axial stress decreases, as can be seen in equation (5). Also, it is notable that axial stress decreases as relative density decreases in equation (6). Therefore, the type II process, where the inner and outer compacts are combined after precompaction, was carried out. The average relative density of the two compacts after precompaction is 0.45. Figure 10b shows the calculated relative density distributions for the type II process. The distribution of relative density shows the highest density at the top of the core part and the lowest density at the bottom of the core. However, the difference in relative density between minimum and maximum values in type II is 0.026, which is smaller than that in the type I process. Also no height difference between inner core and outer ring is observed in Fig. 10b. The reduced density difference and no height difference shows that the friction force decreases when the average relative density before combination decreases.

Effects of clearance between inner and outer compacts during combination (types II and III)
In order to reduce the friction force technically, the type III process was used. Here, the important technical point is that the inner diameter of the upper ring punch is smaller than the diameter of the lower core punch to make a designed clearance between the inner and outer precompacts before combination. It is expected that the inner precompact is inserted into the outer precompact without contact, if the clearance between inner and outer precompacts is big enough for elastic expansion of the precompacts. If the clearance is not big enough to obtain no contact, the radial stress in the
precompacts will be reduced and eventually the friction force on contact surfaces will also be reduced. Figure 13 shows the initial finite mesh used for the type III process. Figure 10 shows calculated relative density distributions for the type III process. The difference in between minimum and maximum relative density is 0.014, which is a more even distribution of density than that in the type II process. Figure 14 shows the top and bottom surfaces of a duplex compact made by the type III process. It is notable that there occurs no height difference between the inner and outer compacts, which represents a small friction force during combination. Figure 15 shows the relative density distribution calculated by FEM for the case of a large inner diameter, 6.8 mm, and that measured experimentally, respectively. The density distribution calculated by FEM show an even distribution throughout the duplex compact and agrees considerably well with the experimental results.

CONCLUSION

Three types of duplex compaction process have been simulated by FEM using a backward Euler method and a yield criterion to describe the densification behaviour of ceramic powder. Two parameters in the yield criterion for powder compaction could be determined from the uniaxial compaction pressure versus relative density data. The friction force on the contact surfaces during combination of inner and outer compacts plays an important role in making the density heterogeneous and the height difference. The friction force can be largely reduced in the type III duplex compaction process by giving a clearance between the inner and outer compacts during combination. Experiments also have been carried out using a ZrO₂ powder. The height difference between the inner and outer compacts has been observed in the type I duplex compact process, however it has not been observed in the type II and III process. Relative density distributions have been measured experimentally for a case of the type III process using an indentation method. The duplex compact made by the type III process shows a more even distribution of relative density than compacts made by other processes, which can also be observed in the calculated results.

APPENDIX

The constants involved in the solution of the elastoplastic constitutive equation are given by

\[
A_{11} = \frac{\partial \Phi}{\partial q} + \Delta \rho \left( B_0 \frac{\partial^2 \Phi}{\partial p \partial q} + \frac{\partial^2 \Phi}{\partial q \partial R} \frac{\partial R}{\partial \Delta \rho} \right)
\]

\[+ \Delta \epsilon ( - 4 R^2 q - \frac{4 R^2 q}{3(1 - \Delta \epsilon p)}) \Delta \rho
\]

\[+ \left[ 6 B_0 (1 - R^2) + \frac{12 R^2 p}{1 + \Delta \epsilon p} \right] \Delta \epsilon (\partial \Phi / \partial p)
\]

\[
A_{12} = \frac{\partial \Phi}{\partial p} + \Delta \rho \left( - 3 G_0 \frac{\partial^2 \Phi}{\partial q \partial q} - \frac{\partial^2 \Phi}{\partial q \partial R} \frac{\partial R}{\partial \Delta \rho} \right)
\]

\[+ \Delta \epsilon ( - 3 G_0 \frac{\partial^2 \Phi}{\partial q \partial q} + \frac{\partial^2 \Phi}{\partial q \partial R} \frac{\partial R}{\partial \Delta \epsilon q}) \Delta \rho
\]

\[= 6(1 - R^2) p - G_0 (4 + 2 R^2) \Delta \epsilon p
\]
\[ A_{21} = B_0 \frac{\partial \Phi}{\partial \rho} + \frac{\partial \Phi}{\partial \rho} \frac{\partial R}{\partial \phi} \frac{\partial \rho}{\partial \phi} \]
\[ = 6B_0(1 - R^2)\rho \]
\[ = - \frac{2}{3} Rq^2 - 6Rq^2 - K^2m \left( \frac{R - R_T}{1 - R_T} \right)^m \frac{1}{1 - R_T} \frac{R}{1 + \Delta \rho} \]
\[ A_{22} = -3G_0 \frac{\partial \Phi}{\partial \rho} + \frac{\partial \Phi}{\partial \rho} \frac{\partial R}{\partial \phi} \frac{\partial \rho}{\partial \phi} \]
\[ = -G_0(4 + 2R^2)\rho q \]
\[ b_1 = -\Delta \rho \frac{\partial \Phi}{\partial \rho} - \Delta \rho \frac{\partial \Phi}{\partial \phi} \]
\[ = - \frac{4 + 2R^2}{3} q \Delta \rho - 6(1 - R^2)\rho \Delta \rho \]
\[ A_{22} = - \Phi \]
\[ b_2 = - \Phi \]
\[ = - \frac{2 + R^2}{3} q^2 - 3(1 - R^2)\rho^2 + \left( \frac{R - R_T}{1 - R_T} \right)^m K^2 \]

REFERENCES