Numerical Simulation of Microstructure Evolution of Heavy Steel Casting in Casting and Heat Treatment Processes

Bin SU,1) Zhiqiang HAN,1)* Yongrang ZHAO,2) Bingzhen SHEN,2) Enxian XU,2) Shujun HUANG2) and Baicheng LIU1)

1) Key Laboratory for Advanced Materials Processing Technology (Ministry of Education), School of Materials Science and Engineering, Tsinghua University, Beijing, 100084 China. 2) CITIC Heavy Industries Co., Ltd., Henan, 471039 China.

(Received on August 29, 2013; accepted on November 11, 2013)

The microstructure evolution of hypoeutectoid steel during casting and heat treatment processes was simulated by using cellular automaton method. In the simulation, the peritectic solidification, α phase precipitation and pearlite transformation during casting process were considered, and the austenite formation, grain coarsening and decomposition during heat treatment were simulated. The final microstructure, including the average grain size and fraction of α phase as well as the average interlamellar spacing of pearlite, was obtained. The use of through-process simulation as well as a comparison with experiments was demonstrated using a hollow shaft casting as an example. By using the model, the microstructure evolution at different locations in the hollow shaft was simulated, in which the thermal history data obtained by simulating the casting and heat treatment processes were adopted. Metallographic samples taken from the test bar were examined and corresponding mechanical properties tests were conducted. The simulated results were compared with the experimental results to validate the model.

KEY WORDS: cast steel; casting and heat treatment processes; numerical simulation; microstructure evolution; cellular automaton method.

1. Introduction

Large and heavy castings are widely used in the manufacturing of equipments for metallurgy, shipbuilding, and electric power industries. It is very important to improve the performance and quality of heavy castings. In particular, microstructure evolution is unique at a given position inside the castings because it depends mainly upon thermal history which can be quite different in a casting with large size and different wall thickness. Therefore, it is of great significance to study the phase transformation kinetics and the microstructure evolution in steels during casting and heat treatment processes.

Modeling and simulation play an important role in improving casting quality, reducing leading time and cost.1) In recent years, cellular automaton (CA) method has been successfully applied to simulate the microstructure evolution of steel, and it can provide not only the general microstructure information, such as volume fraction of phases, average grain size and overall transformation kinetics, but also the actual microstructure evolution, such as the morphology of phases, distribution of grains and solute concentration.2–7) Kang et al.2) used a CA model to simulate the microstructure evolution of steel ingot during solidification process, in which the columnar to equiaxed transition (CET) was considered. Yang et al.3) used a CA model to simulate the austenitization of hypoeutectoid steel. In their models, the dissolution of pearlite and the transformation of ferrite into austenite were simulated. Li et al.4,5) developed a CA model to simulate the microstructure evolution of the transformation of austenite into ferrite. In their models, the morphology of ferrite, distribution of grains and solute concentration evolution were simulated. However, most of the research work focuses on the microstructure evolution in one process, i.e., either casting process or heat treatment process separately, the effect of the initial microstructure on the subsequent phase transformation, such as the effect of as-cast microstructure on austenitization and the effect of austenite microstructure on austenite decomposition, was usually neglected. In the actual production, large and heavy castings usually undergo casting and heat treatment processes. So it is necessary to develop a model to simulate the through-process microstructure evolution during casting and heat treatment processes to establish a numerical approach to get well understanding on the microstructure evolution in the whole material processing chain.

The aim of the present work is to develop a numerical model to simulate the microstructure evolution of a hypoeutectoid steel during casting and heat treatment processes. The purpose of microstructure simulation during casting process is to predict the as-cast microstructure, which also provided the necessary input information for microstructure simulation of heat treatment process. The developed models were applied to simulate the microstructure evolution of the hollow shaft of a ball mill from industry. The temperature
field during casting and heat treatment processes was simulated for obtaining the thermal history of different locations, which was used for microstructure simulation. This paper demonstrates a numerical approach to conduct through-process modeling of microstructure evolution during casting and heat treatment processes.

2. Model Description

2.1. Phase Transformation during Casting Process

The microstructure evolution of hypoeutectoid steel during solidification and consequent cooling process includes the molten metal transforms into ϒ-ferrite, peritectic transformation, and the transformation of austenite into α-ferrite and pearlite.6)

When \( T_{L}<T<T_{k} \) (\( T_{L} \) is liquidus temperature, \( T_{k} \) is peritectic temperature), molten metal transforms into \( \delta \)-ferrite. A continuous nucleation model \(^{6} \) was employed to calculate the nucleus number in the undercooled melts

\[
\frac{dn_{\delta}}{d(\Delta T)} = \frac{N_{\text{max}}}{\sqrt{2\pi\Delta T_{\alpha}}} \exp\left(-\frac{1}{2}\left(\frac{\Delta T - \Delta T_{\alpha}}{\Delta T_{\alpha}}\right)^{2}\right) \quad \text{...... (2)}
\]

where \( \Delta T \) is the undercooling, \( n_{\delta}(\Delta T) \) is the nucleus density, \( N_{\text{max}} \) is the maximum nucleus density, \( \Delta T_{\alpha} \) is the standard deviation of the distribution, \( \Delta T_{\alpha} \) is the mean nucleation undercooling, and \( f_{3}(\Delta T') \) is the fraction of solid phase. The increment of solid fraction was calculated based on the relationship between solid fraction and temperature that was obtained using Thermo-Calc software,\(^{9} \) where Scheil model was employed. The growth velocity of \( \delta/L \) interface was evaluated using the calculated increment of solid fraction based on the assumption of spherical grain growth.

Peritectic reaction, \( L+\delta \rightarrow \gamma \) (L is molten metal, \( \delta \) is \( \delta \)-ferrite, \( \gamma \) is austenite), takes place when the temperature reaches \( T_{k} \), which is followed by the growth of austenite as a consequence of the transformations of \( \delta \) to \( \gamma \) and L to \( \gamma \). It is assumed that the austenite covers the surface of a \( \delta \) grain immediately. The growth velocity of \( \gamma/L \) interface was calculated also using the relationship between solid fraction and temperature. A mixed-mode model \(^{10} \) was employed to calculate the growth velocity of the \( \gamma/\delta \) interface, where both the finite interface mobility and the finite diffusivity of the alloying elements are taken into account.

\[
v^{1/6} = M^{1/6} \Delta G_{\delta \rightarrow \gamma} \quad \text{...... (3)}
\]

where \( v^{1/6} \) is the interface growth velocity, \( M^{1/6} \) is the interface mobility, and \( \Delta G_{\delta \rightarrow \gamma} \) is the chemical driving force.

\( \alpha \)-ferrite starts to nucleate at the \( \gamma \) grain boundaries when the temperature drops to \( A_{1} \). The nucleation rate, \( I \), can be expressed as \(^{11} \)

\[
I = \frac{2D_{C}V_{x}}{a^{4}(3kT)^{1/2}} \exp\left(-\frac{\Delta G_{\alpha}^{2}}{kT}\right) \quad \text{...... (4)}
\]

where \( D_{C} \) is the diffusion coefficient of carbon in \( \gamma \), \( V_{x} \) is the atomic volume of iron, \( x \) is the atomic fraction of carbon, \( a \) is the average lattice parameter of \( \alpha \) and \( \gamma \), \( \eta \) and \( \xi \) represent parameters depending on the potential nucleation site density and interfacial energies, \( k \) is Boltzmann constant, and \( \Delta G_{\alpha} \) is the driving force for \( \alpha \) nucleation. The nucleation density for \( \alpha \) phase, \( n_{\alpha} \), can be expressed as

\[
n_{\alpha} = \frac{\int_{A}^{1} I(T') \phi(T')dT'}{(\phi(T'))^{n}} \quad \text{...... (5)}
\]

where \( \phi(T') \) is the cooling rate. The growth kinetics of the \( \alpha/\gamma \) interface was determined by using the mixed-mode model as well.

The residual austenite transforms to pearlite instantly at a temperature lower than \( A_{1} \). Scheil’s additivity rule was employed to determine the transformation temperature during the continuous cooling condition, where the criterion is that the sum of the fractional incubation time for all temperatures below \( A_{1} \) equals to 1.\(^{12,13} \) In this study, the incubation period of isothermal transformation, \( t_{\text{inc}} \), is a function of temperature

\[
t_{\text{inc}} = \frac{A \exp\left(\frac{Q}{RT}\right)}{(\Delta T)^{m}} \quad \text{...... (6)}
\]

where \( A \) and \( m \) are parameters associated with steel grade and cooling condition. \( Q \) is activation energy for carbon diffusion. The interlamellar spacing of pearlite, \( S_{p} \), can be calculated by\(^{14} \)

\[
S_{p} = \frac{4c\sigma V_{m}}{\Delta G_{\gamma \rightarrow \gamma}} \quad \text{...... (7)}
\]

where \( c \) is a parameter, \( \sigma \) is interface energy, \( V_{m} \) is molar volume of pearlite, \( \Delta G_{\gamma \rightarrow \gamma} \) is the driving force for pearlite transformation.

2.2. Phase Transformation during Heat Treatment Process

The microstructure evolution of hypoeutectoid steel during heat treatment can be divided into three stages: austenite formation, grain coarsening and decomposition.

Austenite formation in hypoeutectoid steel occurs in two steps: the dissolution of pearlite and the transformation of \( \alpha \)-ferrite into austenite. A continuous nucleation model \(^{7,15} \) was employed to calculate the nucleus number of austenite in the pearlite region

\[
n_{\gamma} (\Delta T) = f_{\gamma} \exp\left(\frac{-Q_{u}}{k\Delta T}\right) \quad \text{...... (8)}
\]

where \( \Delta T \) is the value of overheating (\( \Delta T = T - A_{c1} \)), \( n_{\gamma}(\Delta T) \) is the nucleus density, \( f_{\gamma} \) is the factor representing the influence of pearlite structure on the nucleation, \( Q_{u} \) is the activation energy of nucleation, and \( k \) is Boltzmann’s constant. Austenite grains grow into the pearlite region at a velocity \( v^{p} \).

\[
v^{p} (\Delta T) = f_{p} \exp\left(\frac{-Q_{G}}{k\Delta T}\right) \quad \text{...... (9)}
\]

where \( f_{p} \) is the factor representing the influence of pearlite structure on the growth rates, and \( Q_{G} \) is the activation energy of growth. The mixed-mode model was employed to calculate the growth velocity of the \( \gamma/\alpha \) interface.

After the full austenite microstructure is formed, austenite
interface migration distance. In the simulation, the effect identifying variable, (2) cell status, (3) concentration, and square cells. Each cell has the following variables, (1) grain
transition rules also determine the state of each cell based
ermed and updated by the transition rules. Meanwhile, the
model. At each time step, the variables of each cell are gov-
growth kinetics and was used in cell capturing in the CA
interface migration distance was calculated based on the
tations were obtained by solving diffusion equations. The
of carbon and manganese were considered and their concen-
2.3. Mechanical Properties Prediction
Plenty of the microstructure-property study has been car-
ceed on low-carbon ferrite-pearlite steels, in which the
gin size and fraction of α-ferrite and interlamellar spacing
pearlite determine the mechanical properties. The form of
the equation is

\[ \sigma_y = f_a \sigma_y + \left(1 - f_a \right) \sigma_p \] .......................... (12)

where \( \sigma_y \) is yield strength or tensile strength of ferrite-pearlite aggregate, \( \sigma_y \) is yield strength or tensile strength of ferrite, \( \sigma_p \) is yield strength or tensile strength of pearlite, and \( f_a \) is the fraction of ferrite. The strength of ferrite and pearlite can be predicted by the well-known Hall-Petch relationship

\[ \sigma_a = \sigma_y + k_d^{-1/2} \] .......................... (13)

\[ \sigma_p = \sigma_y + k_S^{-1/2} \] .......................... (14)

where \( \sigma_a \) and \( \sigma_p \) are the friction stress, \( k \) and \( k_S \) are the Hall-
Petch parameters, \( d \) is grain size of ferrite and \( S \) is inter-
lar spacing of pearlite. By substitution from Eqs. (13) and
(14) into Eq. (12), \( \sigma_y \) is given by

\[ \sigma_y = f_a \left( \sigma_y + k_d^{-1/2} \right) + \left(1 - f_a \right) \left( \sigma_y + k_S^{-1/2} \right) \] .... (15)

The HB Hardness can be determined from the tensile
strength (\( \sigma_{UTS} \)) by the linear equation with constants \( a \), \( b \)

\[ HB = -a + b \sigma_{UTS} \] .......................... (16)

3. Model Validation
CA method was used in the simulation. A two-
dimensional computational domain was discretized into
square cells. Each cell has the following variables, (1) grain
identifying variable, (2) cell status, (3) concentration, and
(4) interface migration distance. In the simulation, the effect
carbon and manganese were considered and their concen-
trations were obtained by solving diffusion equations. The
interface migration distance was calculated based on the
growth kinetics and was used in cell capturing in the CA
model. At each time step, the variables of each cell are gov-
ermed and updated by the transition rules. Meanwhile, the
transition rules also determine the state of each cell based
on the previous state of the cell and the states of its neigh-
boring cells.
In our previous work, the microstructure evolution of
ASTM A216 WCA cast steel during solidification and con-
sequent cooling process was simulated using the developed
model. To validate the model, a step-shaped sand-mold cast-
ing was produced to investigate the influence of cooling rate
on the as-cast microstructure in the steel plant of CITIC
Heavy Industries Co., Ltd.. Thermocouples were positioned
at the center of the steps to measure the temperature varia-
tion during the solidification process of the casting. Samples
for microstructure observation were taken from the center of
the steps. It was shown that the simulated results were in
good agreement with the experimental results.
In the subsequent work, the microstructure evolution of
ASTM A216 WCA cast steel during heating process was
simulated using the developed model. To validate the mod-
el, dilatometric and quenching experiments were carried out
and experimental samples were taken from the center of the
Step 1. The dilatometric experiment was conducted using a
DIL805A dilatometer, and experimental data was employed
to study phase transformation kinetics and validate the mod-
ule. While the quenching experiment was conducted using a
chamber electric furnace, and metallographic examination
was carried out. The simulated results were compared with
the experimental results and the capability of the model for
quantitatively predicting the microstructure evolution of the
steel in heating process was assessed.
A series of experiments was carried out to develop micro-
structure-property relationships for continuously cooled
hypoeutectoid steel. Samples were heated to different soaking
temperatures and cooled with different cooling rates.
Microstructure analysis was conducted to determine the
size and fraction of α-ferrite and interlamellar spacing
of pearlite quantitatively in steel and corresponding
mechanical properties tests were conducted. The equations
for the mechanical properties have been developed by uti-
lizing data from both the laboratory and the literature

\[ \sigma_{YS} (\text{MPa}) = f_a \left( 77.6 + 14.4d^{-1/2} \right) + \left(1 - f_a \right) \left( 305.9 + 92.2\omega(\text{Mn}) + 0.32S^{-1/2} \right) + 0.79\omega(\text{Si}) \] .... (17)

\[ \sigma_{UTS} (\text{MPa}) = f_a \left( 122 + 15.9d^{-1/2} \right) + \left(1 - f_a \right) \left( 517 + 0.791S^{-1/2} \right) + 500\omega(\text{Si}) \] .... (18)

\[ HB = -195.1 + 0.626\sigma_{UTS} \] .......................... (19)

where \( \sigma_{YS} \) is yield strength, \( \sigma_{UTS} \) is tensile strength, \( \omega(\text{Mn}) \)
is the manganese content, wt.%, and \( \omega(\text{Si}) \) is the silicon
content, wt.%. 

4. Application to the Hollow Shaft of a Ball Mill
The hollow shaft of a ball mill (Fig. 1), which produced
in the steel plant of CITIC Heavy Industries Co., Ltd., was
investigated using the modeling approach. Several locations
were selected to investigate the microstructure evolution at
different locations in the casting. The geometry of the cast-
ing and three different locations inside the casting are shown
in Fig. 2. The radius and height of the casting are 1106 mm
and 2,675 mm, respectively. The composition of the steel is listed in Table 1, and the pouring temperature was about 1,554°C. The hollow shaft was produced by sand casting and normalizing treatment was carried out after shake-out and clearing. The sketch of heating temperature versus time is shown in Fig. 3. Thermocouples were positioned at the center of the test bars to measure the temperature variation during casting and heat treatment processes.

Metallographic samples taken from the center of test bars were polished and then etched with 4% nital for microstructure observation by optical microscope and scanning electron microscope (SEM). The corresponding mechanical properties were determined by the tensile tests and the hardness tests. The tensile testing was carried out using standard

| Table 1. The chemical composition of the steel (wt.%). |
|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| C   | Si   | Mn   | S   | P   | Ni   | Cr   | Cu   | Mo   |
| 0.26 | 0.36 | 0.78 | 0.01 | 0.012 | 0.08 | 0.07 | 0.1 | 0.05 |

Fig. 1. The hollow shaft of a ball mill.

Fig. 2. The geometry of the hollow shaft (a) and its longitudinal section (b). A, B and C are three different locations selected to investigate the microstructure evolution inside the casting.

Fig. 3. The sketch of heating temperature versus time.

Fig. 4. The microstructure of samples taken from the center of the test bars. (a) as-cast microstructure, (b) final microstructure after heat treatment, (c) the morphology of lamellar pearlite of as-cast microstructure, and (d) the morphology of lamellar pearlite of final microstructure.
specimens following the international standard ISO 6892-1:2009. The hardness tests are performed on polished samples by the hardness tester TH320. The simulated results were compared with the experimental results to validate the model. Figure 4 shows the as-cast microstructure and final microstructure of samples taken from the center of test bars.

The flow chart of the simulation process is shown in Fig. 5. The casting process was simulated by using FT-Star software we have developed, and the macroscopic temperature variation in heat treatment process was simulated by using SysWeld software. The simulated results of the temperature field are shown in Fig. 6. Figure 7(a) shows the cooling curves of the selected locations inside the casting during solidification process. By using the cooling curves and the developed model, the microstructure evolution during casting process can be simulated, and the as-cast microstructure can be predicted to provide the necessary input information for microstructure simulation of heat treatment process. Figure 7(b) shows the temperature change curves of the selected locations during heat treatment process. The selected locations inside the casting during casting and heat treatment processes are the same ones. By using the temperature change curves and the developed model, the microstructure evolution during heat treatment process can be simulated, and the final microstructure and mechanical properties can be predicted.

As we all know, the determination of nucleation parameters in microstructure simulation is difficult. Usually, these parameters are determined by comparing numerical results with experimental observations. In our work, we referred to the parameters provided in literatures2–5,12–17) and utilized our own experimental data to calibrate our model parameters. These parameters were given in our previously published papers.6,7)

The simulated microstructure evolution at the center of the test bar (Point A) during solidification and consequent cooling process is shown in Fig. 8. The computational domain is divided into 300×200 rectangular cells. The cell size is 5×5 μm. Figure 8(a) shows the evolution of δ-ferrite precipitating from the liquid, where the green area represents δ-ferrite and the red area represents the liquid metal. Figure 8(b) shows peritectic transformation process, where the gray area represents the austenite phase. Figure 8(c) shows the simulated as-cast microstructure, where the gray area represents the pearlite while all the color area rep-
It can be seen that the morphology and grain size of the simulated microstructure are quite close to the observed result (Fig. 4(a)). The simulated results of as-cast microstructure at different locations inside the casting are shown in Table 2. Because of the different cooling rates, the microstructure is nonuniform in the casting. For a faster cooling rate, the average grain size of α-ferrite and the average interlamellar spacing of pearlite are smaller.

The simulated microstructure evolution at the center of the test bar (Point A) during heating process is shown in Fig. 8. The simulated microstructure evolution at the center of the test bar (Point A) during heating process, where P is pearlite, γ is austenite and α is α-ferrite. (a) 730°C, (b) 810°C, (c) 10 hours at 900°C, and (d) 30 hours at 900°C.

The simulated austenite microstructure at Point B (a) and Point C (b) after heating process (30 hours at 900°C) is shown in Fig. 10.

The simulated microstructure evolution at the center of the test bar (Point A) during cooling process. (a) 730°C, (b) 710°C, and (c) room temperature. The simulated final microstructure at Point B (a) and Point C (b) after cooling process is shown in Fig. 12.
Table 2. The simulated results of as-cast microstructure at different locations inside the casting.

<table>
<thead>
<tr>
<th>As-cast microstructure</th>
<th>Point A</th>
<th>Point B</th>
<th>Point C</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Simulation</td>
<td>Experiment</td>
<td>Simulation</td>
</tr>
<tr>
<td>$d_{as}$, $\mu$m</td>
<td>182.5</td>
<td>188.2</td>
<td>232.3</td>
</tr>
<tr>
<td>$S_p$, $\mu$m</td>
<td>0.392</td>
<td>0.421</td>
<td>0.62</td>
</tr>
<tr>
<td>$f_o$</td>
<td>0.638</td>
<td>0.652</td>
<td>0.615</td>
</tr>
</tbody>
</table>

Table 3. The simulated results of final microstructure after heat treatment at different locations inside the casting.

<table>
<thead>
<tr>
<th>Final microstructure</th>
<th>Point A</th>
<th>Point B</th>
<th>Point C</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Simulation</td>
<td>Experiment</td>
<td>Simulation</td>
</tr>
<tr>
<td>$d_{as}$, $\mu$m</td>
<td>27.2</td>
<td>26.5</td>
<td>34.5</td>
</tr>
<tr>
<td>$S_p$, $\mu$m</td>
<td>0.34</td>
<td>0.31</td>
<td>0.362</td>
</tr>
<tr>
<td>$f_o$</td>
<td>0.596</td>
<td>0.59</td>
<td>0.607</td>
</tr>
</tbody>
</table>

Table 4. The predicted mechanical properties of the final microstructure at different locations inside the casting.

<table>
<thead>
<tr>
<th>Mechanical properties</th>
<th>Point A</th>
<th>Point B</th>
<th>Point C</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{UTS}$, MPa</td>
<td>536</td>
<td>541.8</td>
<td>525</td>
</tr>
<tr>
<td>$\sigma_{YS}$, MPa</td>
<td>286</td>
<td>290</td>
<td>277</td>
</tr>
<tr>
<td>HB</td>
<td>140.4</td>
<td>148.1</td>
<td>133.5</td>
</tr>
</tbody>
</table>

9. Figure 9(a) shows the transformation of pearlite into austenite, where the black area represents pearlite and the red area represents ferrite. The other color areas represent $\gamma$ phase, and different colors represent $\gamma$ grains with different crystallographic orientations. Figure 9(b) shows the transformation of ferrite into austenite. Figures 9(c) and 9(d) show austenite grain coarsening process. The simulated austenite microstructure at Point B and Point C after heating process is shown in Fig. 10. Figures 9(d) and 10 are used as the initial austenite microstructure for austenite decomposition during cooling process. It can be seen that the initial austenite microstructure are different at different locations inside the casting because of the difference of the as-cast microstructure and the thermal history.

The simulated microstructure evolution at the center of the test bar (Point A) during cooling process is shown in Fig. 11. Different orientation ferrite grains are represented by different colors, and the initial austenite is represented by black area. From Fig. 11, we can see that ferrite grains nucleate at austenite grain boundaries and then grow into austenite grain interior. It can be seen that the simulated microstructure are quite close to the observed result (Fig. 4(b)). The simulated final microstructure at Point B and Point C after cooling process is shown in Fig. 12.

The simulated results of final microstructure after heat treatment at different locations inside the casting are shown in Table 3. With the results of the microstructure, mechanical properties can be calculated from Eqs. (17)–(19). The predicted mechanical properties of the final microstructure at different locations inside the casting after heat treatment are shown in Table 4. The results show that mechanical properties of the hollow shaft casting can meet the expectant requirements basically, and casting technological design and heat treatment process design are reasonable.

5. Conclusion

In this paper, a numerical approach was demonstrated to realize the through-process modeling of microstructure evolution during casting and heat treatment processes. The microstructure evolution of hypoeutectoid steel during casting and heat treatment processes was simulated by using cellular automaton method. In the simulation, peritectic solidification, $\alpha$ phase precipitation and pearlite transformation during casting process were considered, and the austenite formation, grain coarsening and decomposition during heat treatment were simulated. The final microstructure, including the average grain size and fraction of $\alpha$ phase as well as the average interlamellar spacing of pearlite, was obtained, and mechanical properties were predicted. The developed model has been applied to the hollow shaft of a ball mill from industry. This makes it possible to investigate microstructure evolution at different locations inside the casting and optimize the casting and heat treatment processes.

Acknowledgement

This work is funded by the National Science and Technology Major Project of China (2011ZX04014-052 and 2012ZX04012011).

REFERENCES