Electric current-driven migration of electrically neutral particles in liquids

Journal Item

How to cite:

Zhang, Xinfang and Qin, Rongshan (2014). Electric current-driven migration of electrically neutral particles in liquids. Applied Physics Letters, 104(11), article no. 114106.

Version: Accepted Manuscript

Link(s) to article on publisher's website:
http://dx.doi.org/doi:10.1063/1.4869465

Applied Physics Letters **104**, 114106 (2014); doi: 10.1063/1.4869465

Electric current-driven migration of electrically neutral particles in liquids

Xinfang Zhang* and Rongshan Qin*

Department of Materials, Imperial College London, Exhibition Road, London SW7 2AZ,
United Kingdom

Abstract

We design and experimentally demonstrate a migration of electrically neutral particles in liquids driven by electric current according to the discrepancies of their electrical conductivities. A force from electric current to electrically neutral particles has been identified to drive the particles toward the lateral surface from the centre of suspension via three distinguishable zones, namely pushing, trapping and expelling zones. The driving force can overtake gravity in practical cases. The property of the force is found neither similar to that of the force in electromagnetophoresis nor similar to that of the electromigration force in terms of direction and magnitude. An expression for the force at the pushing zone has been developed based on the numerical calculation of the thermodynamics of suspension fluids. The excellent agreement between numerical calculations and experimental data demonstrates that our calculation provides fundamental and predictive insight into particles separation from the liquids. Therefore, it is possible to use the force in many engineering applications such as separation of particles according to the differences of their electrical conductivities.

*Corresponding authors. Tel.: +44 (0)20 7594 6803; fax: +44 (0)20 7594 6757.

E-mail addresses: xin-fang.zhang@imperial.ac.uk (X.F. Zhang), r.qin@imperial.ac.uk (R.S. Qin).

Much attention has been paid to the migration of electrically neutral particles from the liquids due to the increasing demands for separation technique. Electrically neutral particles can migrate in a magnetic field traversed by an electric current. The migration is perpendicular to the current and to the homogenous magnetic field that is maintained at right angles to the current. It is Lorentz force that causes migration of particles. Normally, this magnetic field-induced migration of particles in liquids is called electromagnetophoresis.[1-4] However, if the applied external magnetic field is removed, how does the electric current affect the movement of the neutral particles? It has been known that the electric current causes electromigration.[5-8] The force in the electromigration comes from the momentum transfer from the conducting electrons to atoms. The direction of electromigration force is opposite to that of the electric field. This force will not contribute to the movement of particles in the direction perpendicular to the electric current. Previously it has been established that many inclusions, e.g. oxides, sulfides and gas bubbles, have lower electrical conductivities than that of the metal matrix. When the dimension of an inclusion is significant to the dimension of the matrix, the current distribution is sensitive to the location of the inclusion in the suspension system. In general, different current distributions correspond to different system free energies.[9] In this Letter, a possible separation mechanism to use electric current to expel a high resistivity object from a low resistivity matrix has been proposed based on experimental observations and numerical calculations. This provides a possibility to use electric current to manipulate the transport of electrically neutral non-metallic inclusions in a conductive liquid, which would be of great interest and of physical importance if this kind of migration was beneficial to the separation of particles of nearly equal density but distinctly different electrical conductivity. Therefore, the technique is useful to many practical cases such as galvanizing, coating, welding and purification, where inclusions in a thin layer of liquid metal film need to be calibrated. It is also applicable for the separation of particles such as cells of different tissues, algae, bacteria, and possibly viruses.

Here, pulsed electric current,[10-14] as an instantaneous high energy input method with high efficiency and low energy consumption, is applied to the molten steel containing the MnS inclusions[15] to manipulate the transport of electrically neutral non-metallic inclusions in a conductive liquid. The steel containing inclusions is selected for study due to the particles' position driven by electric current can be recorded in the cooled cast steel. However, the liquids at room temperature do usually not have such characteristics. To compare the distribution of the particles with and without electric current treatment, one set of liquid was solidification without electric current treatment but with exactly the same thermal treatment history. Another set of sample was treated with the electric current. The pulse is in square wave to reduce the skin effect. Each pulse has a loading duration of 60 μs, current density of 1.6×10^6 A/m^2. The total treatment time is 10 minutes. The frequency of the pulse during the treatment was 1 Hz. The sample is in a width of about 22 mm. The consumed electric power in electric current pulse processing is 0.0012 Watts, which is much less than the power of household fluorescent. In order to confirm the effect from electric current, the above-mentioned experiments are performed in the same parameters with mould flux. Mould fluxes are the powders that are fed onto the top of the molten metal surface to absorb particles from the liquid.[16] Detailed experimental procedures are similar to that reported in reference [10]. The cooled samples were longitudinally sectioned and polished for metallographic examination. The distribution of the inclusion across the sample was examined by optical microscope. Numerical calculations have been used to suggest the separation mechanism for the particles from the liquid.

The enumerated data distributions of the MnS inclusions across the samples are as follows. The number of inclusion is distributed almost uniformly in the sample without electric current treatment, but is in a vertical double well shape in the sample treated by electric current (Fig.1). The inclusions in the middle area of the sample are almost pushed away by the electric current. In the area close to the surface, the number of inclusions achieves local maximum. When mould flux is are fed onto the top of the molten metal surface to absorb particles from the liquid, separation of inclusions induced by electric current becomes more significant (Fig.1). The number of inclusions (～25 per unit area) on the surface is three-quarters smaller with respect to that of the sample without mould flux (～100 per unit area). More interestingly, the length of the region that an inclusion being pushed away is widened from 7.9 mm to 16.9 mm in comparison to that of the sample without mould flux. But the distribution of inclusion number keeps unchanged although the mould flux is applied to the molten steel without electric current treatment. This indeed indicates that the electric current drives inclusions in liquid metal to move toward the surface. Once the inclusions reach to the interface between molten steel and mould flux under the driving of the electric current, they will be trapped by the mould flux, accordingly only a few amounts of inclusions present in the electric-current treated steel.

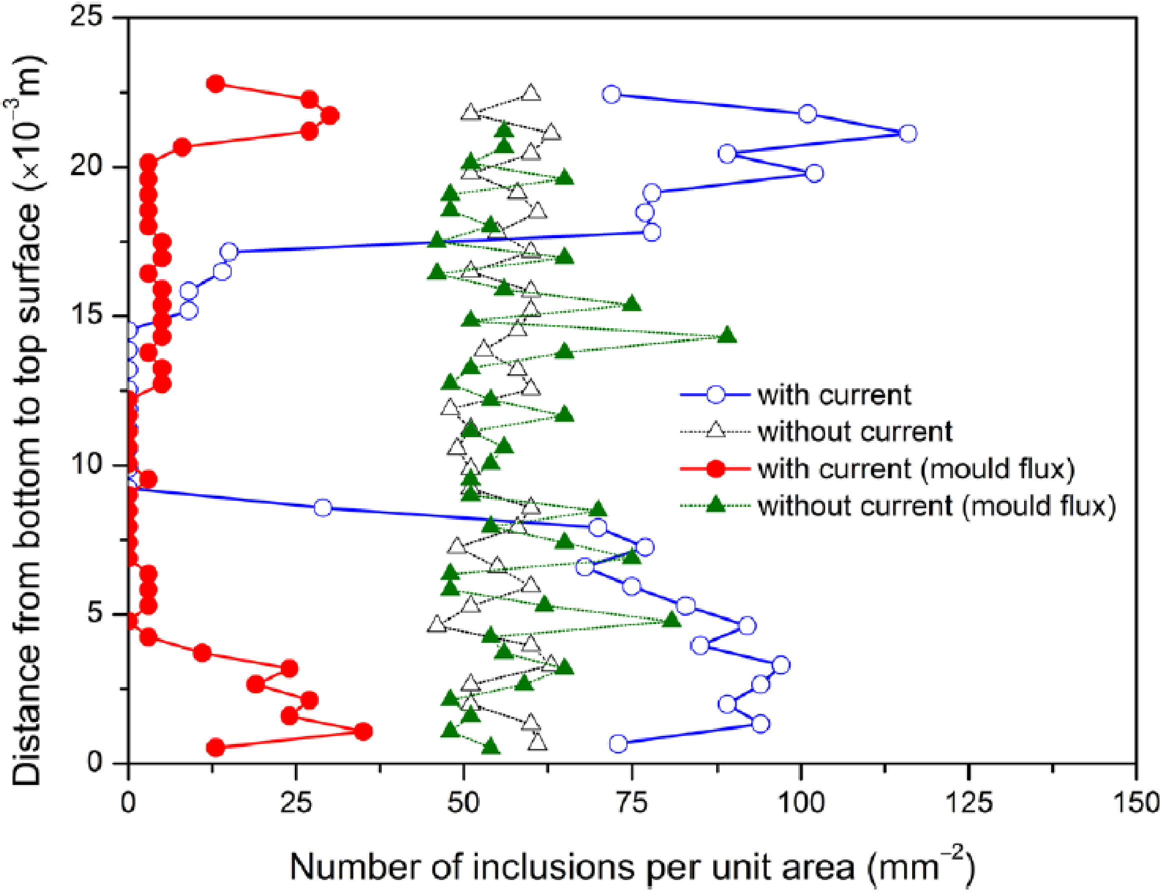


FIG.1. Number distribution of inclusions in liquid metal without and with electric current treatment. The symbols of open circle and hollow triangle represent the MnS inclusions distribution with and without electric current treatment, respectively. It indicates that the inclusions disappear from the inner part of the steel matrix, and instead are dispersed on the surface (e.g. double well shape). The particles are distributed almost uniformly in the sample without electric current treatment. When mould flux is applied, the distribution in the sample without electric current (indicated by solid triangle) still keeps random. The distribution of electric-treated sample with mould flux (indicated by solid circle) exhibits the similar double well, but a smaller number of inclusions.

We have also examined the size distribution of inclusions on further statistical analysis by means of histograms and peaks fitting using Lorentzian. Individual MnS particle with equivalent circular diameter are recorded. The particle under various test conditions has a wide variation in size (4μm<particle size<28μm), but its size distribution mainly concentrates in approximately 8 μm according to Lorentzian fitting (Fig.2). Meanwhile, it is clear that the inclusions smaller than 30 μm can be extracted from molten metals by the electric current. The efficiency for the impurity removal and power consumption (0.0012 Watts) with electric current treatment are far superior to the conventional methods has been reported.[17-20] As mentioned in Fig.1, the inclusions disappear from the inner part of the steel matrix (Figs 2c and 2f), and instead are dispersed at the surface of the steel (Figs 2b and 2e). The uniform distribution of inclusions in the steels without electric current treatment does not suffer dramatic effect regardless of whether the mould flux is used (Figs 2a and 2d).

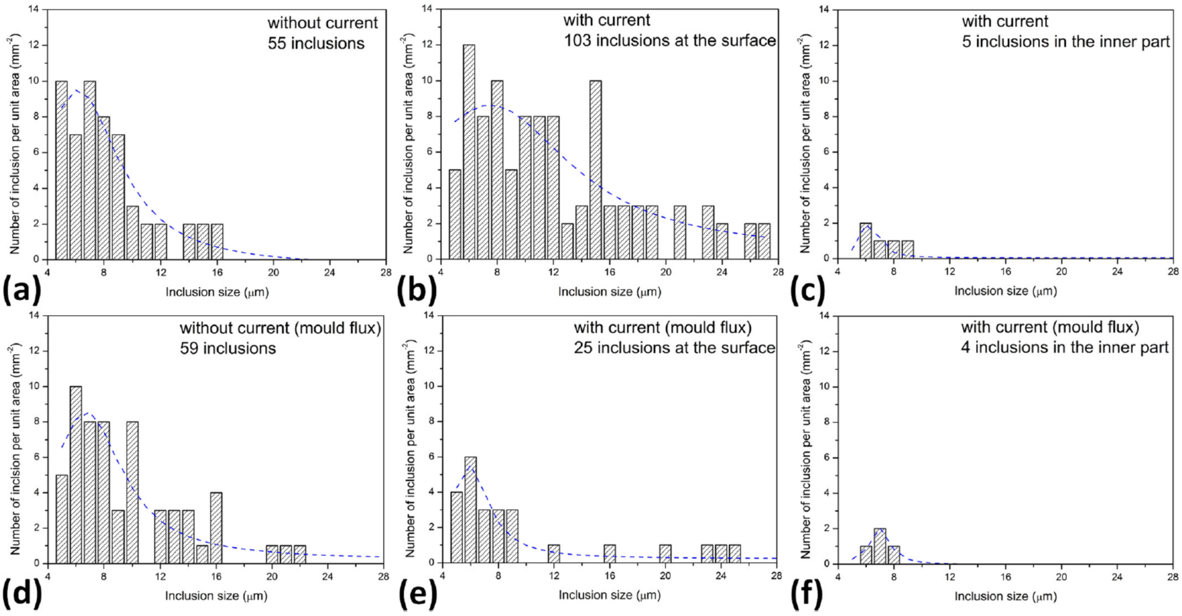


FIG.2. Size distributions of inclusions in liquid metal without and with electric current treatment. Individual MnS particle with equivalent circular diameter are recorded. The peaks are fitted using Lorentzian. (a) Inclusion size in the sample without electric current treatment is in the range of 4 to 28 μm. (b) and (c), After treated by electric current, the inclusions are dispersed at the surface with the varied size of 4 to 28 μm. But they disappear from the inner part of the matrix. (d) The size and number of the particles exhibits the similar trend as in the untreated sample although the mould flux is applied to the steel. (e) and (f), After treated by

electric current with mould flux, the size does not show a large fluctuation, but only a few amount of inclusions can be observed in the sample.

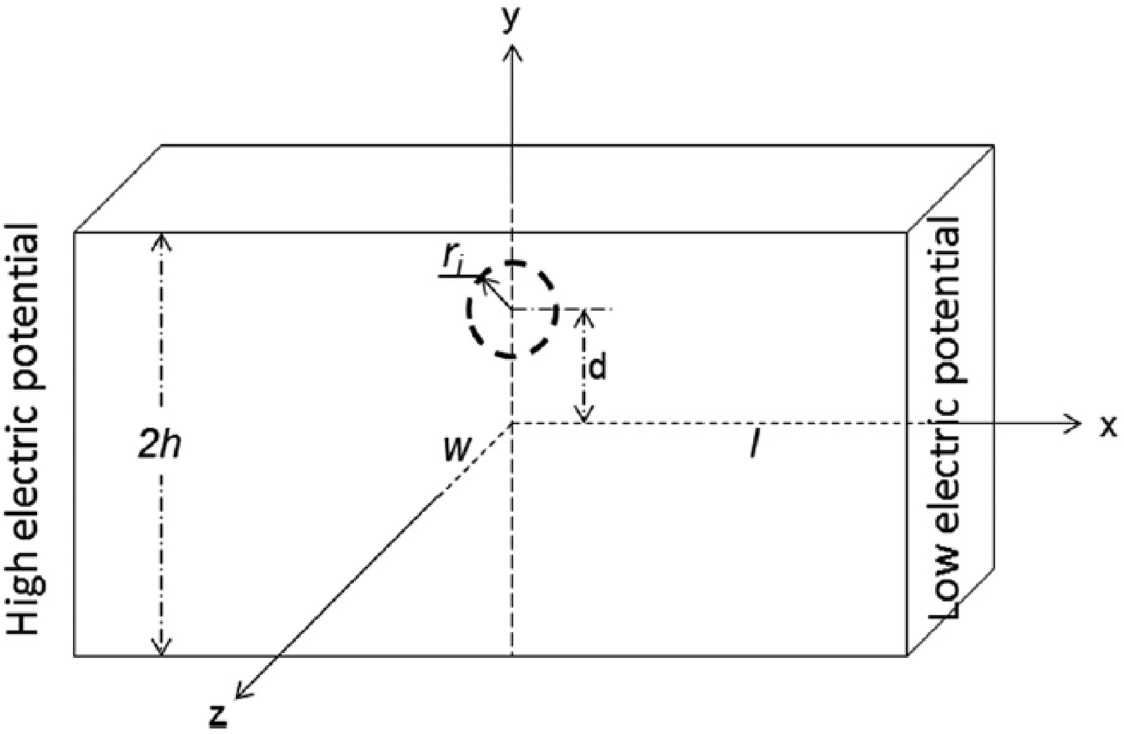


FIG.3. Schematic diagram of the suspension containing a spherical inclusion subjected to an electric potential difference. A spherical inclusion with electrical conductivity σ_i and radius r_i submerged in a rectangular liquid matrix with electrical conductivity σ_m and dimensions $2l \times 2h \times 2w$.

Numerical calculations have been used to suggest the separation mechanism for the particle from the liquid. For simplicity, one considers a spherical particle with electrical conductivity σ_i and radius r_i submerged in a rectangular liquid matrix with electrical conductivity σ_m and dimensions $2l \times 2h \times 2w$. The suspension is subject to an electrical potential difference. The system and the Cartesian coordinates are demonstrated in Fig.3. An electric current is generated in the system due to the electrical potential. As mentioned, the direction of electromigration force induced by electric current is opposite to that of the electric field. In the present work, the force in the perpendicular direction to that of the electromigration is considered. This means that the force discussed in the present work has nothing to do with the conventional electromigration. According to Ohm's law, the electric current density at spatial position *r*, $\vec{j}(r)$, is determined by the local electrical conductivity $\sigma(r)$ and the local electric field $\vec{E}(r)$ via $\vec{j}(r)=\sigma(r)\vec{E}(r)$. $\vec{E}(r)$ is determined by the local electric potential gradient via $\vec{E}(r)=-\nabla\varphi(r)$, where $\varphi(r)$ is the local electrical potential. The distribution of the electric current density is subsequently calculated using $\vec{j}_{km}=\hat{l}_{km}\sigma_{km}(\varphi_m-\varphi_k)/l_{km}$, where l_{km} and $\hat{l}_{km}$ are the length and the unit vector of the element linking lattices *k* and *m*, respectively. The current distribution is substituted to following equation to calculate the electric current free energy G_j [9]

$$G_j=-\frac{\mu}{8\pi}\iint_V\frac{\vec{j}(r)\cdot\vec{j}(r')}{|r-r'|}drdr'+G_j^{ref}, \qquad \textbf{(1)}$$

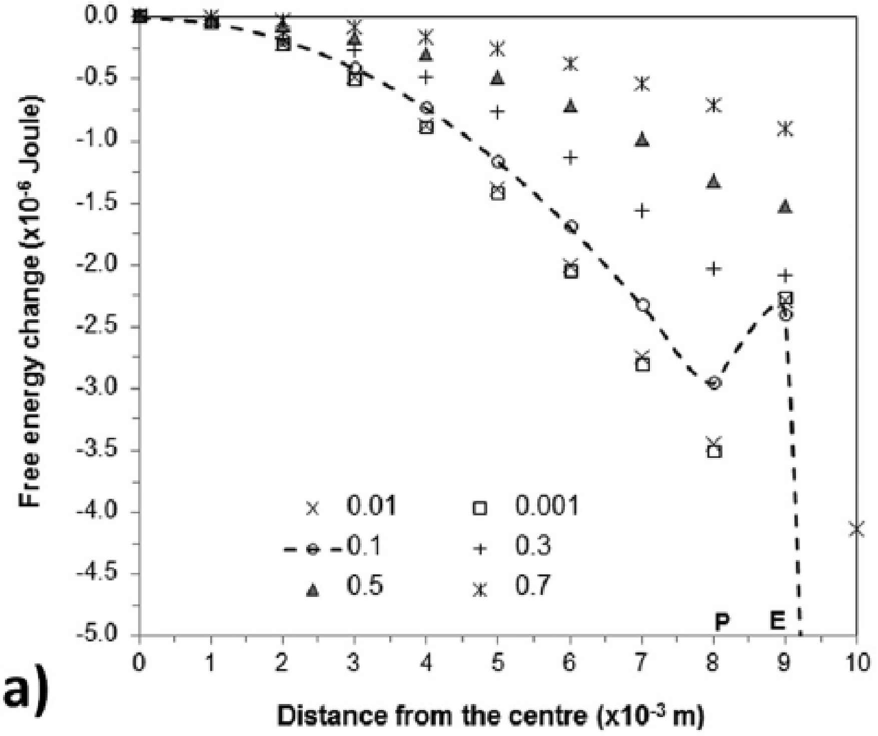
where μ the magnetic permeability, *V* the volume of material, and G_j^{ref} the electric current free energy at the reference state. It should be emphasised that Eq. (1) is the electric current free energy compared to a reference state. The change of the electric current free energy from one set of current distribution to another can be obtained directly using Eq. (1), which is the format appeared in many literatures.[9, 10] The reason to use free energy rather than the free energy change is for the convenience of calculating driving forces in various characteristic lengths (*Δd*). The total system free energy consists of chemical free energy, interface free energy and electric current free energy. When the inclusion moves from one position d to another position $d+\Delta d$, the chemical free energy and interface free energy do not change. The total change of the system free energy equals to the change of the electric current free energy. The equivalent driving force from the electric current to the inclusion in this case is obtained by

$$\vec{F}(d)=-\frac{\partial G_j(d)}{\partial d}=\lim_{\Delta d\to 0}\left[-\frac{G_j(d+\Delta d)-G_j(d)}{\Delta d}\right]. \qquad (2)$$

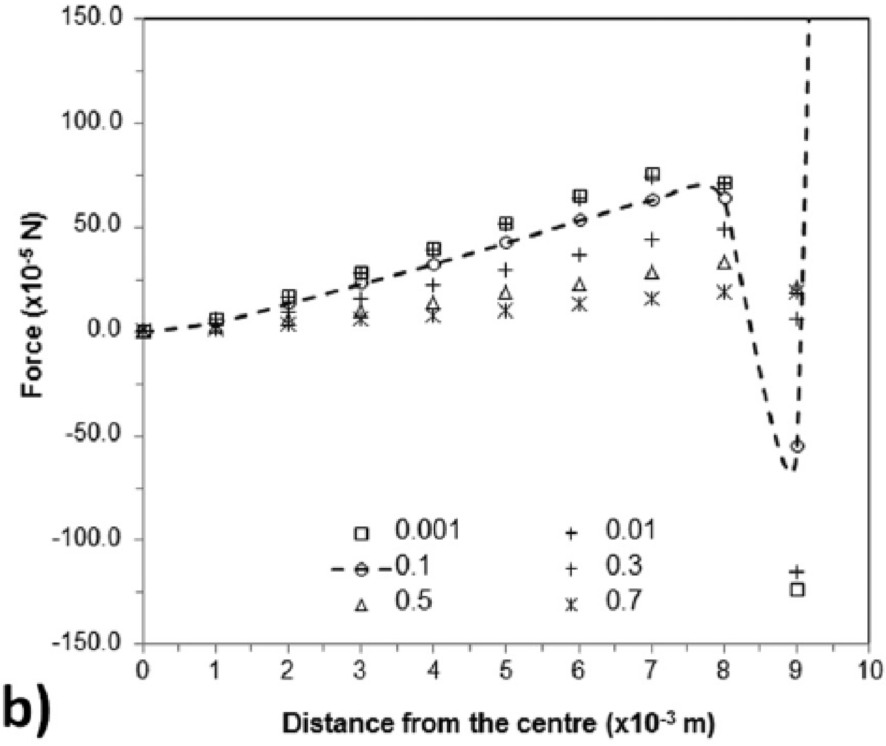
Numerical calculations have been performed to a suspension containing a spherical electrically neutral inclusion. The suspension is subjected to 20 volts electric potential differences. It is also defined that $l=0.1\,m$, $d=0.02\,m$, $w=0.002\,m$, $\sigma_m=1.0\times10^7\,\Omega^{-1}\cdot m^{-1}$

and $\sigma_i = \eta\sigma_m$. Different values of η correspond to different type of inclusions. The value of σ_m is chosen after reference of the electrical conductivity of pure iron at room temperature. The dimensions of the system are chosen after consideration of the sizes of samples used in electric current pulse experiments.[12,14] The distribution of electric current density is then calculated and substituted into Eq. (1) to calculate $G_j(d)$. For convenience, $G_j(0)$ is defined as the reference, where $d = 0$ implies that the inclusion is located at the centre of the matrix. Fig.4a presents the change of the electric current free energy when the inclusion moves from the centre of matrix toward the lateral surface at $r_i = 8.0\times10^{-4}\ m$ and η=0.001, 0.01, 0.1, 0.3, 0.5 and 0.7 respectively. It is found that $G_j(d)$ decreases monotonically from *d=0* to *d=P*. P is a position close to the surface of the matrix. Beyond this point, $G_j(d)$ increases with *d* until $d = E = h - r_i$, where the surface of the inclusion touches the surface of the matrix. Beyond the position E, $G_j(d)$ decreases sharply as the increasing of *d* until the inclusion left the matrix completely. It should be emphasized that $G_j(0)$ is always the largest value. The change of $G_j(d)$ as the increasing of *d* is more severe when η is smaller. However, the differences are not significant when $\eta < 0.01$.

The calculated free energy change has been substituted into Eq. (2) to calculate the equivalent driving force from the electric current to the electrically neutral inclusion. The numerical results are demonstrated in Fig.4b. The force is positive at *0<d<P*, which means the electric current pushes inclusion toward the lateral surface. The area with *0<d<P* is therefore named pushing zone. The force turns to negative at *P<d<E*. This implies that the inclusion will be trapped in the area around position P. The free energy achieves a local minimum at P. The area with *P<d<E* is hence called the trapping zone. The force becomes positive at *d>E* until the inclusion moving out of matrix completely. This area is hence named expelling zone. Electric current helps to remove inclusion from the matrix and also prevent inclusions to enter the metal matrix. To know the strength of the force from electric current to the electrically neutral inclusion, the force-induced acceleration to an inclusion whose mass density is $3.99\times10^{3}\ kg\cdot m^{-3}$ has been calculated. The results are demonstrated in Fig.4c. The mass density is similar to that of MnS at room temperature. It has been found that the accelerations generated by electric current force can exceed the gravitational acceleration in many cases.



(a)



(b)

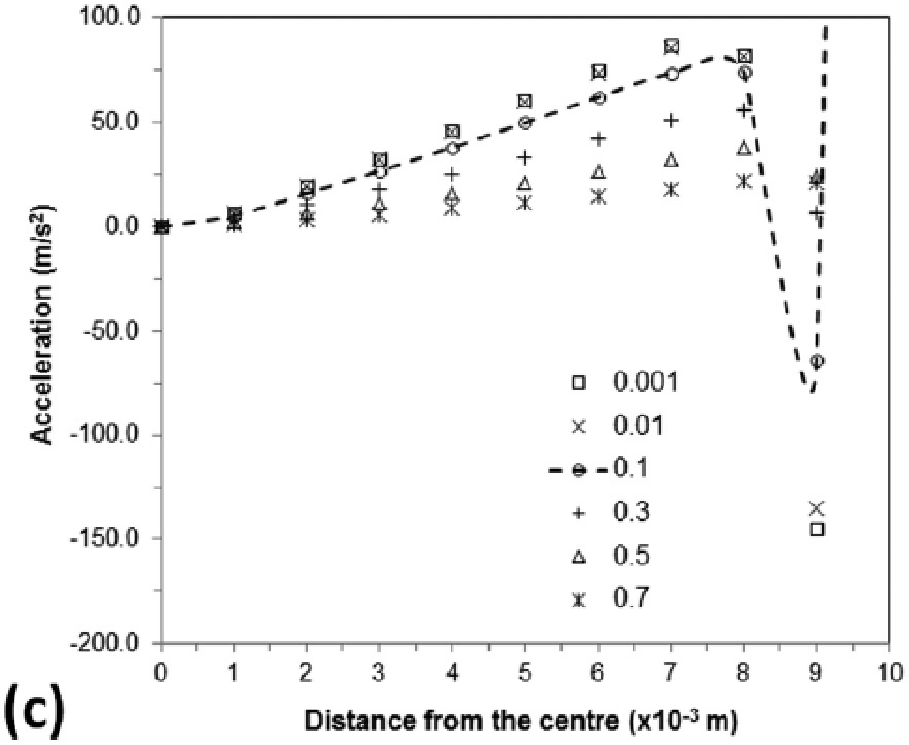
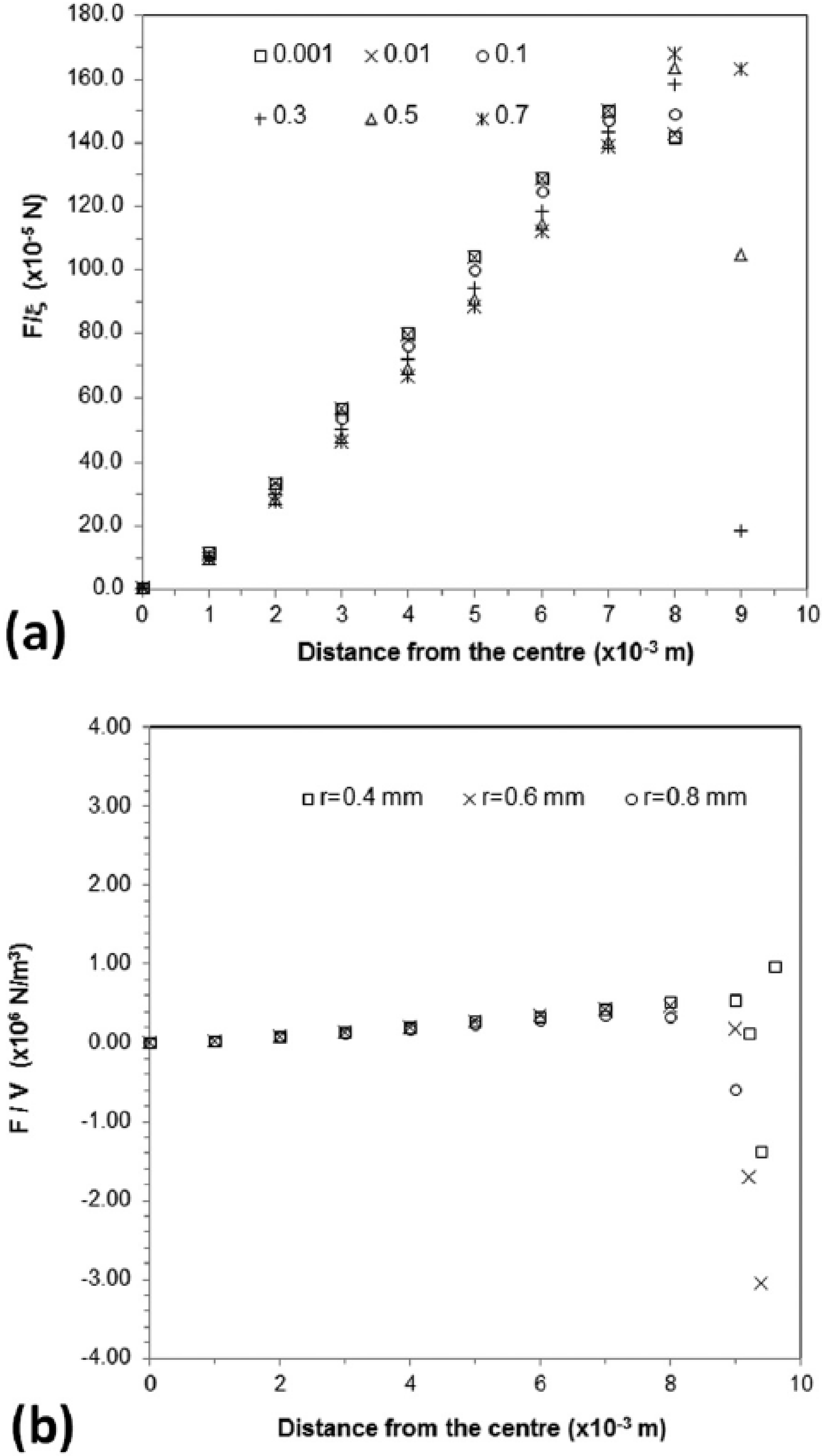


FIG.4. Numerical calculations on the changes of free energy, force and acceleration. **(**a) Free energy change when the inclusion is located at different places. The changes with distance *d* are in the three forms, namely monotonically decreasing (*0<d<P*), monotonically increasing (*P<d<E*) and monotonically decreasing (*d>E*). (b) The force for inclusions at various locations. Three zones are divided according to the nature of the force, namely pushing zone (positive, *0<d<P*), trapping zone (negative, *P<d<E*) and expelling zone (positive, *d>E*). (c) The acceleration caused by the force to a MnS inclusion particle. The driving force can overtake gravity in many cases.

Fig.5a demonstrates the change of F/ξ as a function of d for various η, where $\xi = (\sigma_m - \sigma_i)/(2\sigma_m + \sigma_i) = (1-\eta)/(2+\eta)$. It is found that lines are close to each other in pushing zone (*0<d<P*), although the discrepancies are significant in trapping zone and expelling zone. In approximation, one can say that the force is proportional to the factor ξ in pushing zone. Fig.5b demonstrates the change of F/V_i as a function of d for $\eta = 0.01$ and $r_i = 4.0\times10^{-4}\ m$, $6.0\times10^{-4}\ m$ and $8.0\times10^{-4}\ m$ respectively. It can be concluded that the force is proportional to the volume of inclusion in the pushing zone. Fig.5c shows the change


180.0
160.0
140.0
120.0
100.0
80.0
60.0
40.0
20.0
0.0
F/ξ (x10-5 N)
0.001
0.01
0.1
0.3
0.5
0.7
0 1 2 3 4 5 6 7 8 9 10
(a)
Distance from the centre (x10-3 m)
4.00
3.00
2.00
1.00
0.00
-1.00
-2.00
-3.00
-4.00
F / V (x106 N/m3)
r=0.4 mm
r=0.6 mm
r=0.8 mm
0 2 4 6 8 10
(b)
Distance from the centre (x10-3 m)

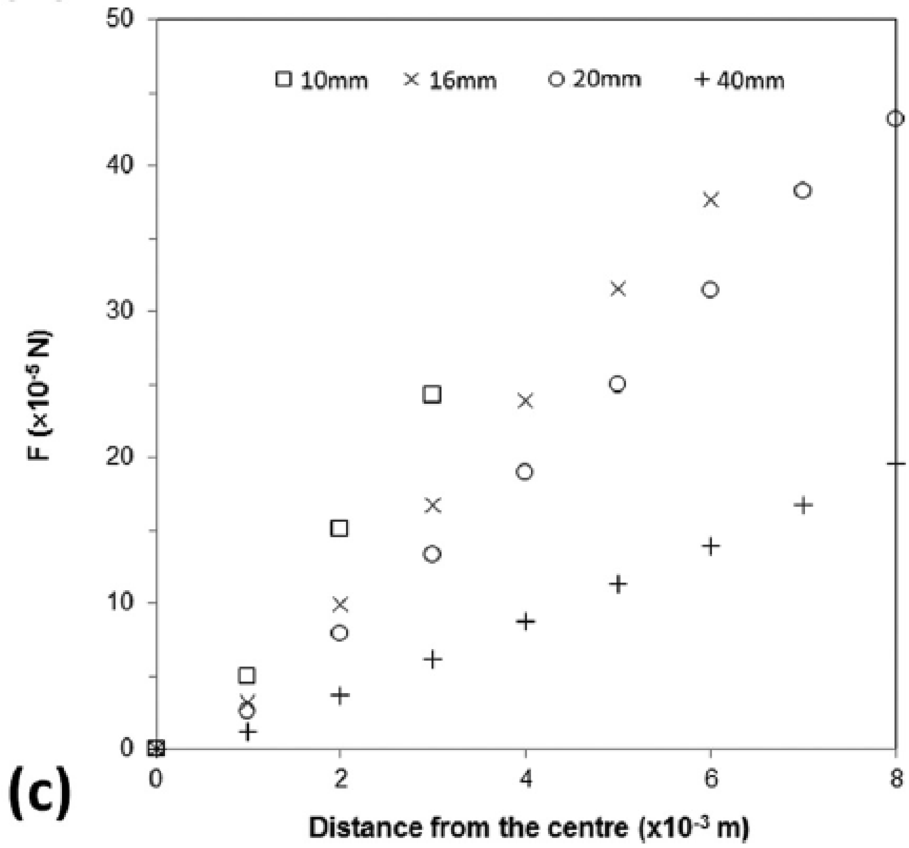


FIG.5. The property of the force from electric current to electrically neutral inclusion. (a) The force for different electrical conductivities of inclusion. The force is proportional to the factor ξ in pushing zone. **(**b) The force for different volumes of inclusion. The force is proportional to the volume of inclusion in the pushing zone. (c) The force for different widths of matrix. The force is proportional to distance for a given width but the slope of the line is dependent on width.

of force as a function of d when $h = 0.01m,\ 0.016\,m,\ 0.02\,m$ and $0.04\,m$ respectively. It can be approximated that F is proportional to d for a given h but the slope of the line is dependent on h. The smaller width of the sample has the steeper slope of the line in Fig.5c. In summary, one has an approximated expression for the force from electric current to the electrically neutral inclusion at pushing zone as

$$F = \mu j_0^2 \frac{d}{f(d)} \frac{\sigma_m - \sigma_i}{2\sigma_m + \sigma_i} V_i = \mu j_0^2 \frac{d}{f(d)} \frac{1-\eta}{2+\eta} V_i, \tag{3}$$

where $f(d)$ increases monotonically but nonlinearly as d increases. The width of the sample is usually much larger than that of the size of inclusion. The pushing zone is, therefore, the dominant area across the matrix in most cases.

The force identified in the present work is different from that in electromagnetophoresis and as followings.[1-4] (a) The direction of the force in electromagnetophoresis is in a fixed direction perpendicular to both the magnetic field and the electric current for $\eta < 1$. While the force in the present work is not in a fixed direction but is from the centre of the matrix to the lateral surface when $\eta < 1$. The latter is axial symmetrical. (b) The amplitude of the force in electromagnetophoresis is not location dependent. While the force revealed in the present work is location dependent. (c) There are three different zones for the force revealed in the present work, namely the pushing zone, the trapping zone and the expelling zone. There are no such zones reported for the force in electromagnetophoresis. (d) The force revealed in the present work does not require any external magnetic force, while the force in electromagnetophoresis requires an external magnetic field in addition of the electric current.

The force is different from that in electromigration with following aspects: (1) The direction of the electromigration force is parallel to the direction of electric current. While the direction of the force identified in this work is perpendicular to that of the electric current. (2) The magnitude of electromigration force to an object is negligible in comparison to its gravitation force. However, the force identified in the present work is significant. (3) The electromigration force to a particle in a matrix can be affected by its surrounding dislocations but will not be affected by its location in the matrix. The force identified in the present work is dependent on the distance from the particle to the surface.

The mechanism proposed, demonstrated and validated here does explain the experimental observation in the study. The disappearance of MnS inclusions from the inner part of the steel matrix and its movement toward the surface is driven by the force from electric current to electrically neutral non-metallic particle. In the area close to the surface, the number of inclusions achieves local maximum (Fig. 1). This shows the trapping effect of electric current to the inclusion at the zone. Further toward the surface, the number of inclusions is reduced. This shows the effect of expelling from the current to the inclusions. Due to absorption of inclusions from mould flux, only a few amounts of inclusions present in the electric-current treated steel than that of without mould flux. The experiments have been repeated for a number of times and the similar results are confirmed.

Electrically neutral particles in liquids may be separated according to the discrepancies of their electrical conductivities. The processing usually requires at least an external magnetic field. This work has identified the existence of a driving force that is caused only by the passing electric current in a finite sample without the application of an external magnetic field. The force drives the electrically neutral inclusions toward the surface. The driving force can overtake gravity in practical cases. The property of the force is found not to be similar to that in electromagnetophoresis and electromigration. Three zones namely the pushing zone, trapping zone and exiling zone are notable for the electric-current-driven phase separation. An expression for the force at the pushing zone has been developed. The theory has been validated by experimental observations on the current-driven separatoin of MnS inclusions from liquid steel. The inclusions (<30 μm) can be extracted from molten metals by the electric current. Therefore, it is possible to use the force in many engineering applications such as separation of particles according to the differences of their electrical conductivities.

The authors are grateful to Prof. Kenneth C. Mills at Imperial College London, Mr Christopher McDonald and Mr Andrew Smith at TATA Steel Teesside Technology Centre for fruitful discussions. The work was financially supported by EPSRC (No. EP/J011460/1), TATA Steel and the Royal Academy of Engineering at United Kingdom.

REFERENCES

[1]A. Kolin, Science **117**, 134 (1953).

[2]H. Watarai, M. Suwa, and Y. Iiguni, Anal. Bioanal. Chem. **378**, 1693 (2004).

[3]D. Leenov, and A. Kolin, J. Chem. Phys. **22**, 683 (1954).

[4]A.H. Latham, R.S. Freitas, P. Schiffer, and M.E. Williams, Anal. Chem. **77**, 5055 (2005).

[5]Y.W. Wang, and P.S. Ho, Appl. Phys. Lett. **103**, 121909 (2013).

[6]X.F. Zhang, J.D. Guo, and J.K. Shang, Scripta Mater. **57**, 513 (2007).

[7]E.C.C. Yeh, W.J. Choi, K.N. Tu, P. Elenius, and H. Balkan, Appl. Phys. Lett. **80**, 580 (2002).

[8]P.S. Ho, and T. Kwok, Rep. Prog. Phys. **52**, 301 (1989).

[9]Y. Dolinsky, and T. Elperlin, Phys. Rev. B **47**, 14778 (1993).

[10]X.F. Zhang, W.J. Lu, and R.S. Qin, Scripta Mater. **69**, 453 (2013).

[11]X.L. Liao, Q.J. Zhai, J. Luo, W.J. Chen, and Y.Y. Gong, Acta Mater. **55**, 3103 (2007).

[12]R.S. Qin, E.I. Samuel, and A. Bhowmik, J. Mater. Sci. **46**, 2838 (2011).

[13]Y.B. Jiang, G.Y. Tang, C.H. Shek, Y.H. Zhu, and Z.H. Xu, Acta Mater. **57**, 4797 (2009).

[14]X.L. Wang, J.D. Guo, Y.M. Wang, X.Y. Wu, and B.Q. Wang, Appl. Phys. Lett. **89**, 061910 (2006).

[15]Q. Meng, G.S. Frankel, H.O. Colijn, and S.H. Goss, Nature **424**, 389 (2003).

[16]K.C. Mills, and A.B. Fox, ISIJ Int. **43**, 1479 (2003).

[17]T. Toh, H. Hasegawa, and H. Harada, ISIJ Int. **41**, 1245 (2001).

[18]L.F. Zhang, J. Aoki, and B.G. Thomas, Metall. Mater. Trans. B **37B**, 360 (2006).

[19]H. Arai, K. Matsumoto, S. Shimasaki, and S. Taniguchi, ISIJ Int. **49**, 965 (2009).

[20]L.F. Zhang, Y.F. Wang, and X.J. Zuo, Metall. Mater. Trans. B **39B**, 534, (2008).