Simulation of crack propagation in tablet film coatings containing pigments

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Summary

A model originally developed to simulate crack propagation in structural steel has been evaluated for tablet film coatings containing pigments. The fracture mechanism is assumed to consist of hole formation around the pigment particles, growth and coalescence. The program allows both the visualisation of crack growth and the calculation of crack velocity. Simulations to investigate the effect of stress intensity factor indicate that the critical stress intensity factor for sustained crack growth in hydroxypropyl methylcellulose is of the order of 0.3 MPa $m^{1/2}$ consistent with experimental findings. Simulations to investigate the effect of the particle size of pigments indicate that the crack velocity is highest at small particle sizes but becomes independent of size above 1–2 μm. In general, agreement with experiment both qualitatively and quantitatively is extremely good.

Introduction

Solid inclusions in the form of aluminium lakes of water-soluble dyes, opacifiers (e.g. titanium dioxide) and various inorganic materials (e.g. iron oxides, calcium carbonate, talc and colloidal silica) are often added to tablet film coatings to improve their colour, opacity, appearance and, in some cases, their film forming characteristics (Rowe, 1984). A potential problem with such materials, highlighted by Porter (1981, 1982), is that of localised cracking around individual particles or aggregates of the solid inclusions. These

defects can have profound effects on the release of an active ingredient especially if the tablet has been coated with an entero-soluble or a sustained release film.

In a comment on this problem, Rowe (1982a) suggested that the effect might be due to differences in the thermal expansion of the solid inclusion and the polymer matrix and he presented literature data on the thermal expansion coefficients for some representative materials to support the hypothesis. The ideas were expanded in a subsequent paper (Rowe, 1986) where it was shown that in the case where the thermal expansion coefficient of a polymer matrix is always higher than that of the solid inclusion (as in the case of pigmented tablet film coatings), the radial and tensile stresses in the polymer matrix at a

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distance r from the centre of solid inclusion of radius R will be $-PR^3/r^3$ and $+PR^3/2r^3$, respectively, where P is given by the expression:

$$P = \frac{\Delta\alpha\,\Delta T}{\dfrac{1+\nu_p}{2E_p} + \dfrac{1-2\nu_i}{E_i}} \tag{1}$$

where E_p and ν_p are the Young's modulus of elasticity and Poisson's ratio, respectively, of the polymer matrix; E_i and ν_i are the Young's modulus of elasticity and Poisson's ratio, respectively, of the solid inclusion; $\Delta\alpha$ is the difference between the coefficient of linear expansion of the polymer matrix and solid inclusion and ΔT is the temperature difference between the processing temperature and room temperature.

From such a treatment, it can be shown that in the case proposed all radial stresses will be compressive and all tangential stresses will be tensile with the result that cracking (if it occurs) will always occur radially from the solid inclusions. From limited data this would appear to be the case for tablet film coatings containing pigments (Porter, 1981, 1982).

A disadvantage of this treatment is that it is concerned only with the static condition and gives no indication of the degree of crack growth and propagation throughout the polymer matrix. In order to investigate these factors it is necessary to use models and computer simulation. One such model, originally developed to simulate crack propagation in structural steels but equally applicable to pigmented tablet film coatings, is that described by Broberg (1990a) and it is this model that has been evaluated in this paper.

The Model

For a detailed description of the model the reader is recommended to consult the papers by Broberg (1989, 1990a). Essential features of the model are:

(a) The inclusions are assumed to be spherical and their diameter determined after a Gauss-ian distribution. The position of the inclusions is randomly distributed.

(b) A hole grows around the inclusions according to a viscosity rule (Broberg, 1989).

$$\frac{1}{R}\cdot\frac{dR}{dt} = \beta\left(\frac{\sigma}{\sigma_0}-1\right)^2\left[1+\left(\frac{\sigma}{\sigma_0}-1\right)^n\right] \tag{2}$$

where σ, the stress, is always greater than σ_0, the flow or yield stress, of the polymer; R represents the radius of the inclusion/hole, t is time, β denotes the fluidity (i.e. the reciprocal of the viscosity) of the polymer and n is an exponent.

(c) The stress is assumed to be scalar and given by the expression

$$\sigma = \frac{K}{\sqrt{\pi(a+\delta)}+\sqrt{\pi a}}$$
$$\times\sqrt{\frac{\sqrt{1-(V^2/C_d^2)\sin^2\theta}+\cos\theta}{1-(V^2/C_d^2)\sin^2\theta}} \tag{3}$$

where K is the stress intensity factor at the crack tip, V represents the crack tip velocity, C_d is the propagation velocity of the irrotational wave, a and θ correspond to the polar coordinates with their origin at the crack tip, and δ is the so-called smoothening length introduced to remove singularity (Broberg, 1990a).

(d) Holes are opened when the stress reaches a critical value dependent on the size of the inclusion.

(e) Hole coalescence occurs when the distance between two holes or one hole and the main crack becomes smaller than a certain critical distance.

(f) Hole opening and growth causes a stress reduction in the neighbourhood of the crack.

The simulation has been written in both QUICK BASIC and Turbo C for a personal computer (Broberg, 1990a,b). The program asks for an input of average inclusion diameter, intended volume fraction, values for the yield stress, fluidity, exponent, irrotational

wave velocity and stress intensity factor for the polymer matrix. Inclusions are displayed on the screen as circles and hole growth by drawing the new periphery for each step. Coalescence between two holes is simply marked by a straight line between the centres.

Experimental

Input data

Unfortunately, accurate values for all the parameters in the equations are not available for the materials commonly used in film coating. However, it is possible to estimate them based on previously published data.

The irrotational wave velocity, C_d, can be calculated using the expression (Broberg, 1990b):

$$C_d = \sqrt{\frac{E_p}{\left(1 - \nu_p^2\right)\rho}} \qquad (4)$$

where E_p is the Young's modulus of elasticity of the polymer (2149 MPa for hydroxypropyl methylcellulose; Gibson et al., 1988), ν_p denotes the Poisson's ratio of the polymer (0.35 for hydroxypropyl methylcellulose; Rowe, 1986), and ρ is the density of the polymer (1.26 g ml^{-3} for hydroxypropyl methylcellulose; Rowe, 1984). Hence, for hydroxypropyl methylcellulose, C_d will be 1390 m^2 s^{-1}.

The fluidity β of polymers will be very high compared to that for steel (1000 s^{-1}; Broberg, 1990a). However, it can be estimated for hydroxypropyl methylcellulose by multiplying the value for steel by the ratio of the Young's modulus of the polymer to that of steel. Hence, β for hydroxypropyl methylcellulose will be of the order of 10^5 s^{-1}.

The yield or flow stress σ_0 can easily be measured using tensile testing or indentation. For hydroxypropyl methylcellulose, σ_0 will be of the order of 30 MPa (Gibson et al., 1988).

The exponent n is related to the sensitivity of the matrix to overstress. For steel, Broberg

(1990a) has assumed a value of 4 but for polymers it will be significantly lower (Broberg, 1990b). For the purposes of this paper n has been taken as unity.

Since one of the objectives of the work is to estimate a value for K, the critical stress intensity factor for sustained crack growth, from simulations, this parameter was left as a variable.

Observations

Crack propagation and velocity

Fig. 1 illustrates a simulation for a system containing 10.4% by volume of an inclusion of mean particle diameter 1 μm assuming a stress intensity factor of 0.3 MPa m$^{1/2}$. In this simulation the crack was programmed to start at point A – in all other simulations the crack has been programmed to start at the edge of the field (see Fig. 4) in order to allow for the formation of more coalescence and cracks necessary for the calculation of crack velocity. The features seen on the simulation – the radial propagation of cracks from the inclusions linking to form a major crack, the tendency for minor cracks to diverge from the main crack (B) and the formation of small cracks independent of the main crack (C) – can be found in tablet film coatings. Fig. 2 shows a photomicrograph of such a system where both a major crack (AB) and a minor branch crack (C) can easily be observed.

It should be noted that any calculations, e.g. for crack velocities and stress intensity factors, from simulations will only be approximate because of the randomised distribution of the particles inherent in the computer program. However, it is possible to gain some insight into the degree of variation by performing repeat simulations with the same input parameters but different randomisations. Fig. 3 shows the results (mean and standard deviations of nine randomisations) for the calculation of crack velocities in a system consisting of 1 μm particles of various volume fractions and two stress intensity factors. In all cases the coefficient of variation for individual results was within the range 15–30%.
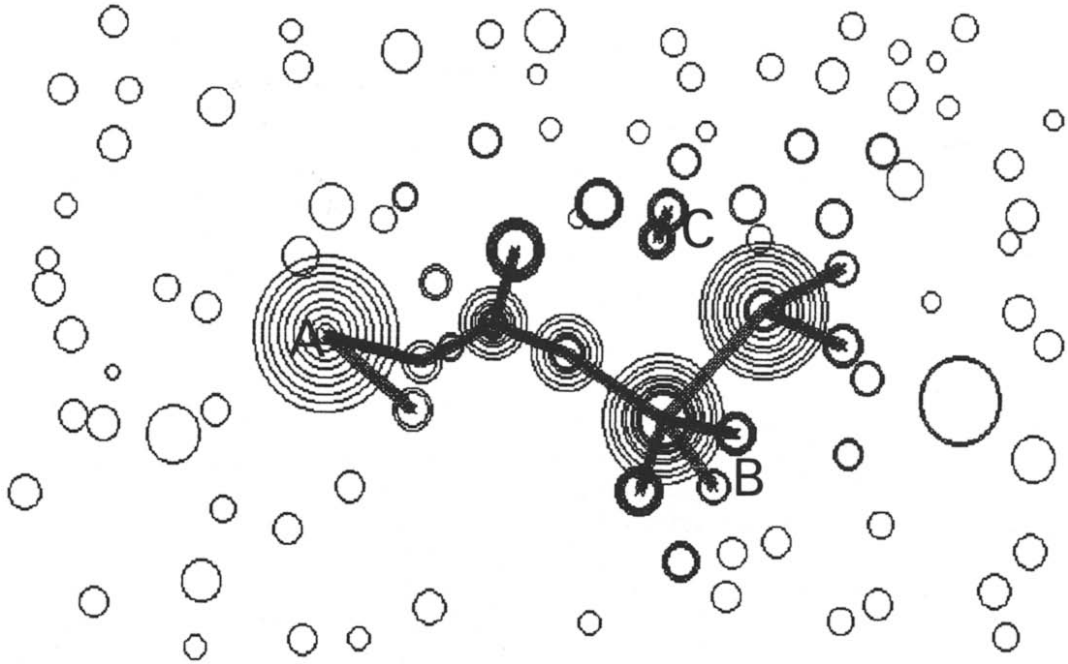
Fig. 1. A simulation of a system containing 10.4% by volume of an inclusion of mean size 1 μm assuming a stress intensity factor of 0.3 MPa m$^{1/2}$. (A) Start of crack; (B) divergent cracks; (C) minor crack.
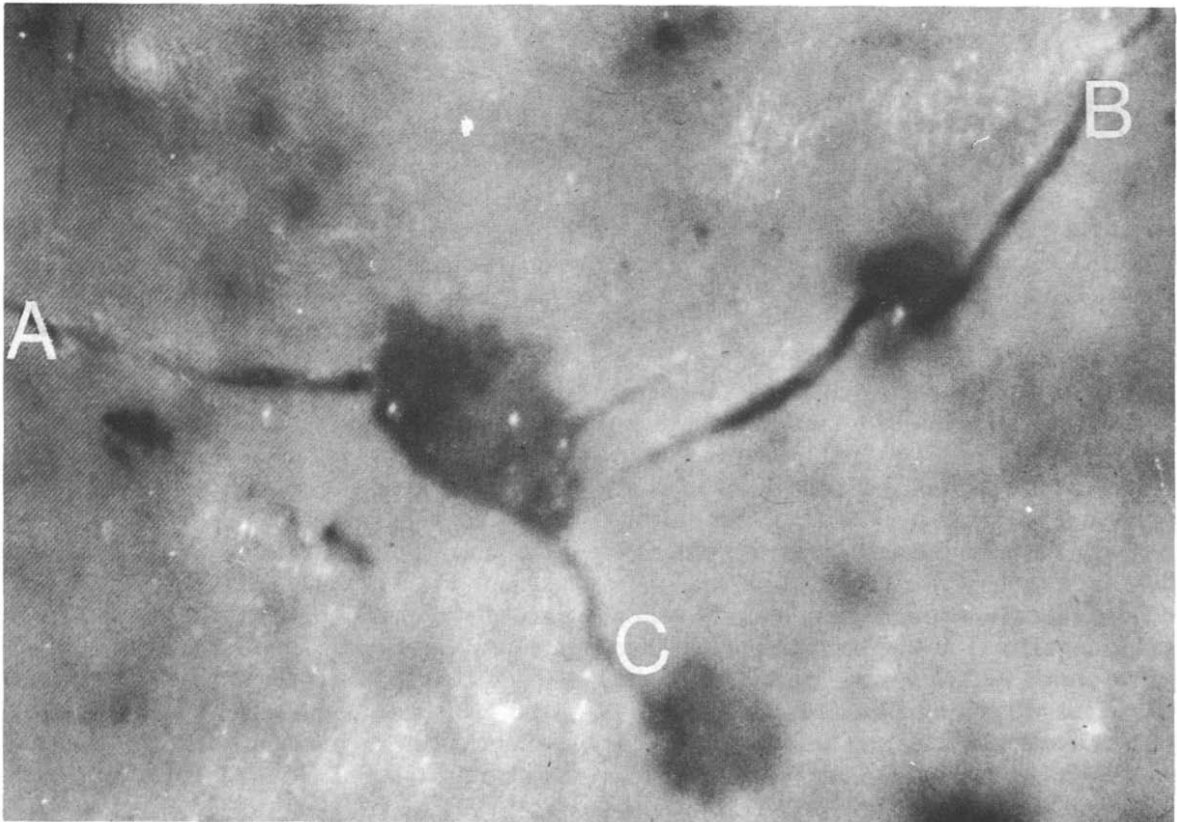

Fig. 2. A photomicrograph showing cracking in a pigmented tablet film coating.
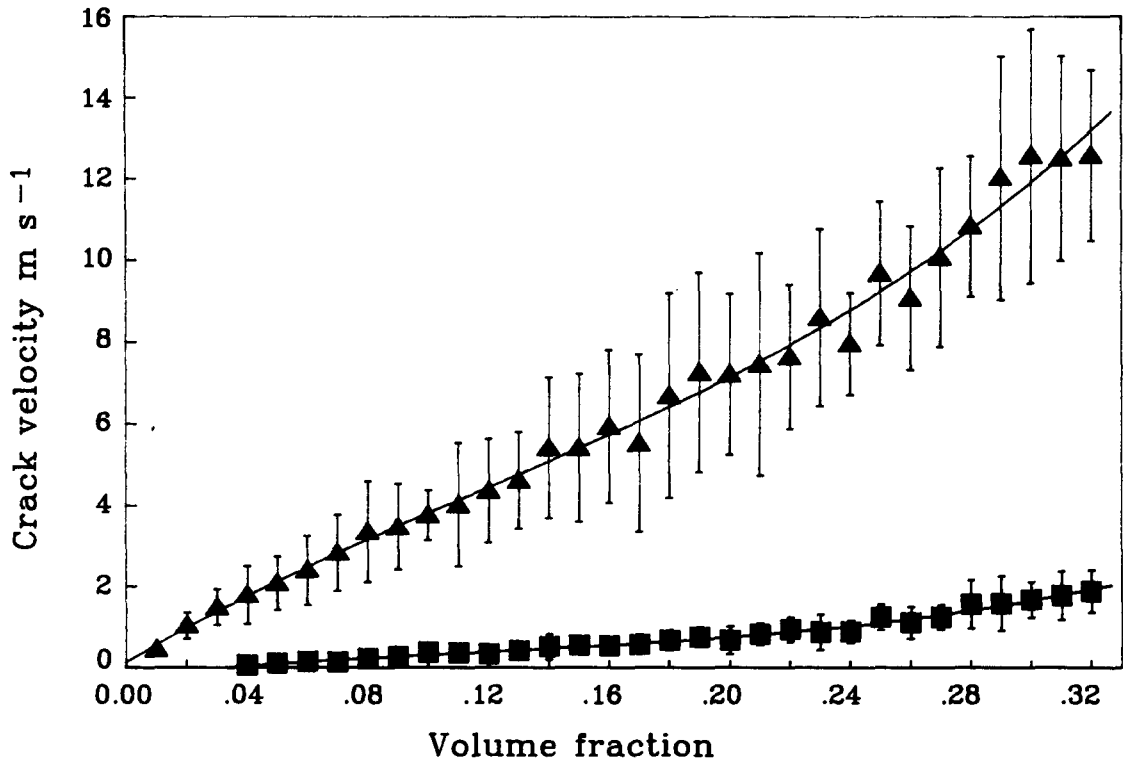
Fig. 3. Effect of volume fraction on crack velocity for a system with two stress intensity factors: 0.3 (■) and 0.5 (▲) MPa m$^{1/2}$.

Estimation of K

Figs 4 and 5 illustrate simulations for a system consisting of inclusions of mean particle diameter of 1 μm at a volume fraction of 0.164 and stress intensity factors of 0.2 and 0.5 MPa m$^{1/2}$, respectively. It is immediately apparent that increasing the stress intensity factor results in the formation of a different crack route, an increase in the number of branched cracks diverging from the main crack, an increase in the number of small cracks independent of the main crack and an increase in crack velocity – the crack velocity at the higher stress intensity factor was 8.72 m s^{-1} compared to 0.26 m s^{-1} for the lower stress intensity factor.

The critical stress intensity factor needed for sustained growth in such a system can be found by performing repeated simulations and pooling the data. Fig. 6 shows such data. It can be seen that for a stress intensity factor of 0.2 MPa m$^{1/2}$ there is no substantial growth below a volume fraction of 0.12 and above this crack velocity is very small. However, at a stress intensity factor of 0.3 MPa m$^{1/2}$ there is sustained crack growth over all the volume fractions studied. There is some dependence of crack velocity on volume fraction but in all cases the velocity does not exceed 2 m s^{-1}. At higher stress intensity factors crack velocity becomes highly dependent on the volume fraction of the inclusions. Hence, it can be concluded that the critical stress intensity factor for this system will be in the region of 0.3 MPa m$^{1/2}$.

Support that this value is not unreasonable for tablet film coatings based on hydroxypropyl methylcellulose can be obtained by using it to predict the stress required to cause catastrophic cracking in these systems. This can be done using the equations recently derived by Brown and Yang (1990) for crack propagation in thin polymer films bonded to rigid substrates. The treatment based on that previously described by Gecit
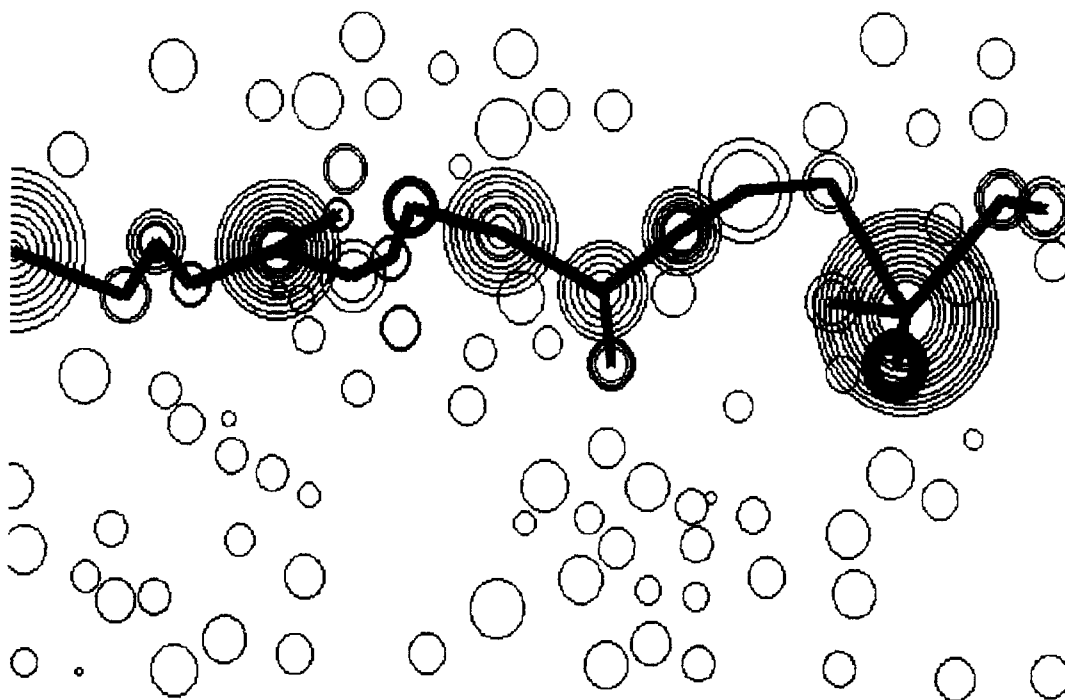
Fig. 4. A simulation of a system contains 16.4% by volume of an inclusion of mean size 1 μm assuming a stress intensity factor of 0.2 MPa m$^{1/2}$.

(1978) predicts that the average strain energy release, G, of a through-thickness crack in a thin film of thickness, h, adhering to a rigid substrate is given by

$$G = 0.36\left(1 - \nu_p^2\right)P^2\pi h/E_p \qquad (5)$$

where P is the internal stress.

Since G is related to the stress intensity factor by the expression

$$G = \left(1 - \nu_p^2\right)K^2/E_p \qquad (6)$$

it can be shown that

$$P^2 = K^2/0.36\pi h \qquad (7)$$

If applied to a tablet coating of thickness 35 μm, P will be of the order of 47.7 MPa. This is very close to the reported tensile strength of hydroxypropyl methylcellulose of 48.9 MPa (Gib-son et al., 1988), indicating that cracking will occur as found in experiment (Rowe, 1981).

Particle size of inclusions

The program is specifically useful for studying the effect of the particle size of the inclusions on crack propagation. Figs 7 and 8 illustrate simulations for two systems with the same volume fraction and stress intensity factor but differing particle sizes of pigments. These form a series with Fig. 5. It should be noted that the figures are at different magnifications, i.e., the program scales the output to include a hundred particles. The effect of decreasing the particle size is similar to the effect of increasing the stress intensity factor, i.e. an increase in the number of cracks diverging from the main crack and an increase in the number of minor cracks independent of the main crack. Pooled data (Fig. 9) show the effect of particle diameter on crack velocity for two stress
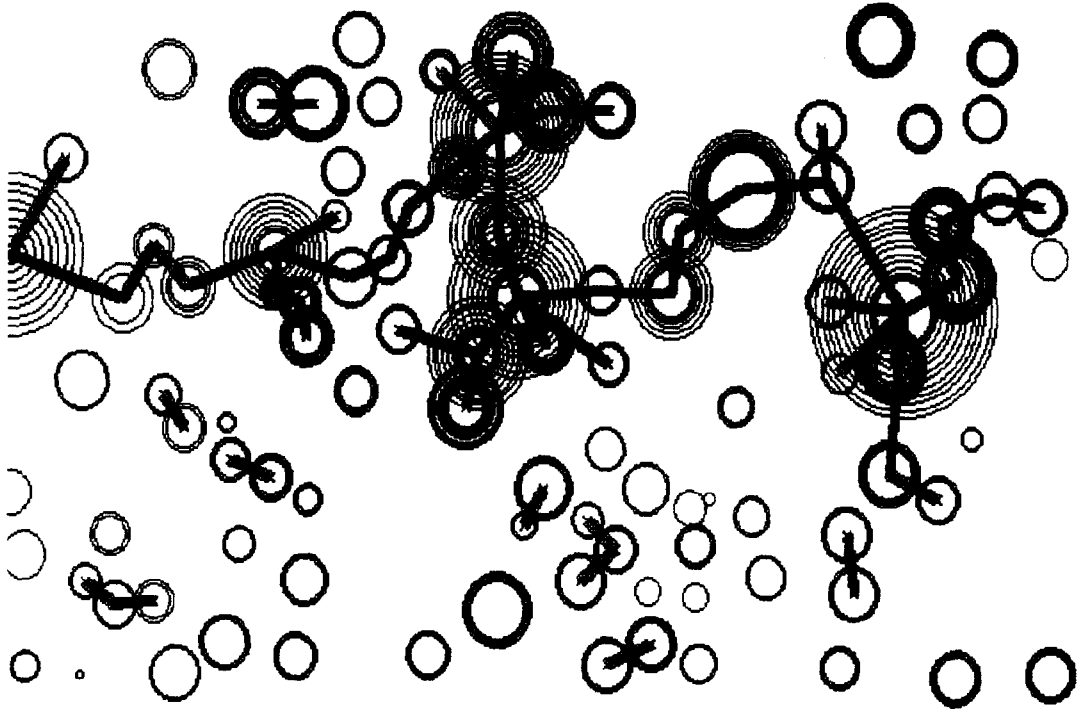
Fig. 5. A simulation of a system containing 16.4% by volume of an inclusion of mean size 1 μm assuming a stress intensity factor of 0.5 MPa m$^{1/2}$.
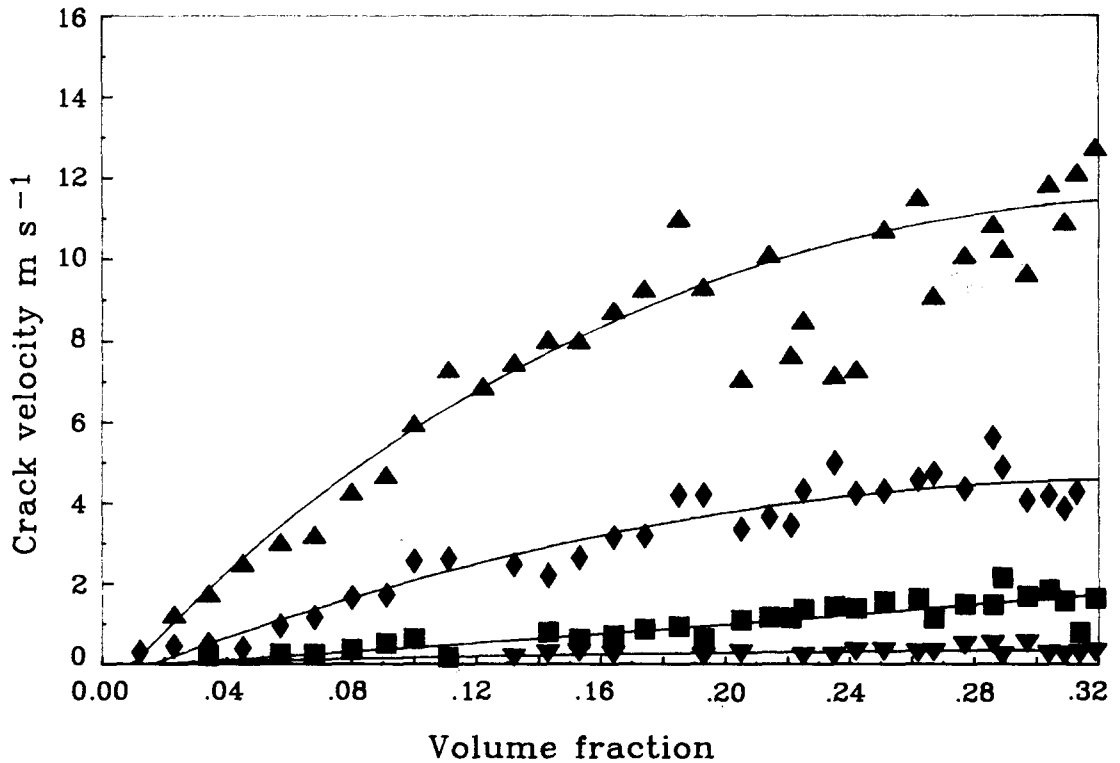

Fig. 6. The effect of volume fraction for a system containing an inclusion of mean size 1 μm on crack velocity for a series of stress intensity factors: ($\blacktriangle$) 0.5 MPa m$^{1/2}$; ($\blacklozenge$) 0.4 MPa m$^{1/2}$; ($\blacksquare$) 0.3 MPa m$^{1/2}$; ($\blacktriangledown$) 0.2 MPa m$^{1/2}$.
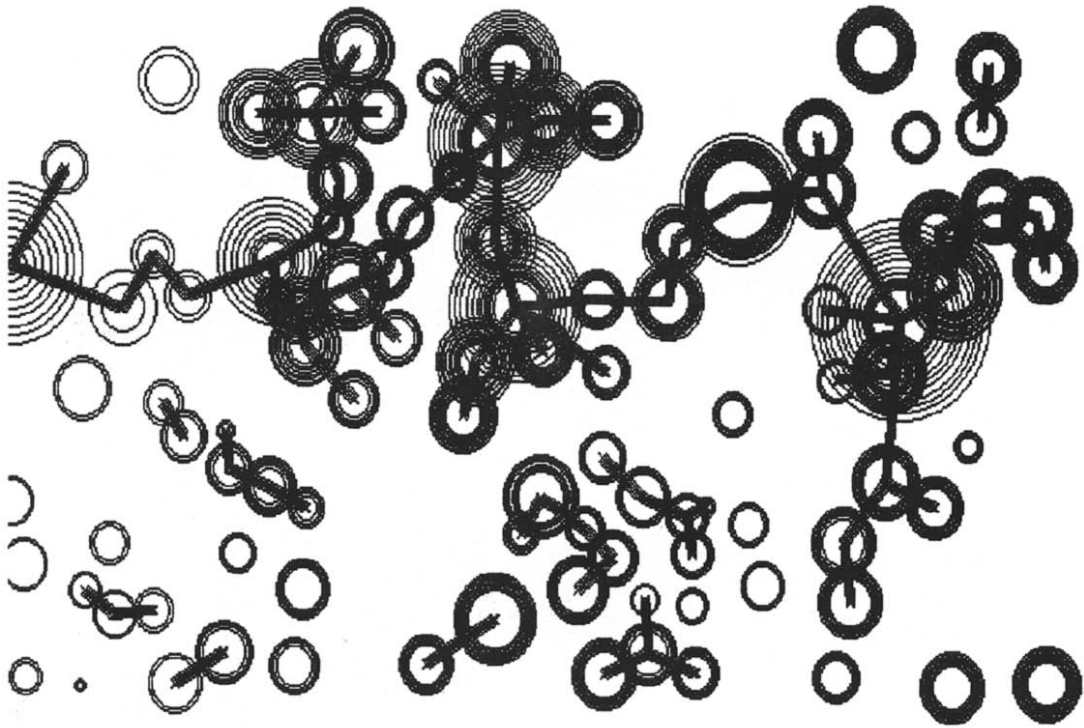
Fig. 7. A simulation of a system containing 16.4% by volume of an inclusion of mean particle diameter 0.25 μm assuming a stress intensity factor of 0.5 MPa m$^{1/2}$.
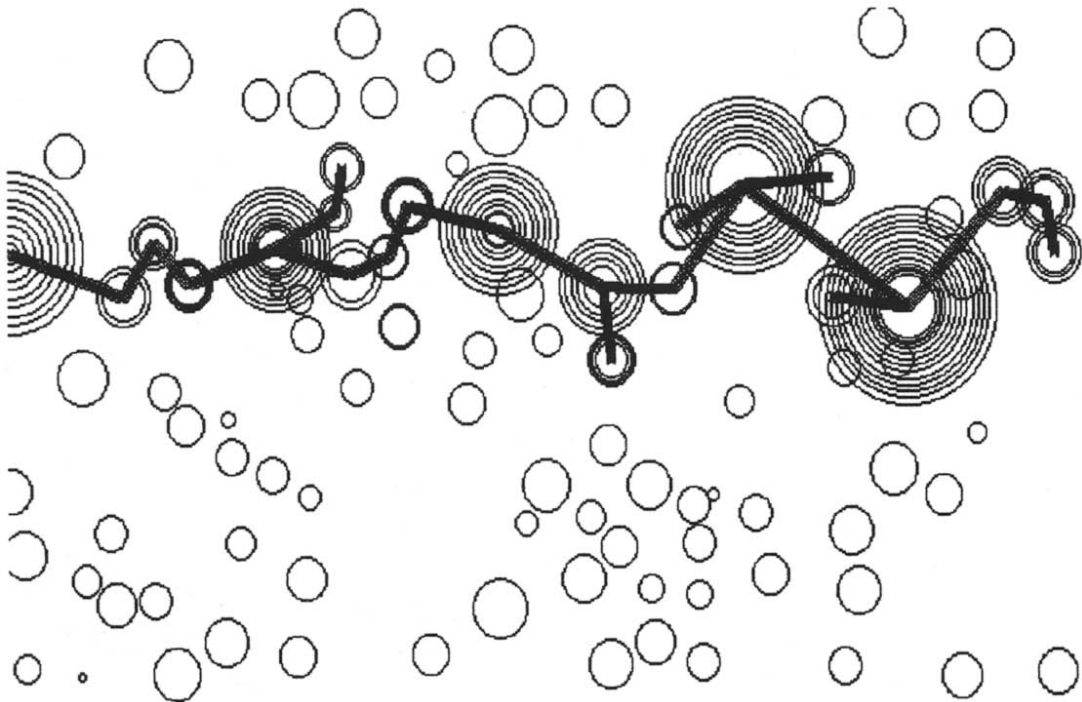

Fig. 8. A simulation of a system containing 16.4% by volume of an inclusion of mean particle diameter 4.0 μm assuming a stress intensity factor of 0.5 MPa m$^{1/2}$.
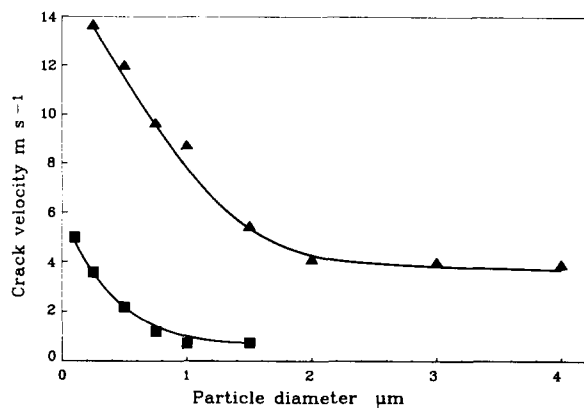
Fig. 9. The effect of particle diameter on the crack velocity of a system containing 16.4% by volume of inclusions assuming two stress intensity factors: (▲) 0.5 MPa $m^{1/2}$; (■) 0.3 MPa $m^{1/2}$.

intensity factors: 0.3 and 0.5 MPa $m^{1/2}$. In both cases, the crack velocity falls dramatically with increasing particle diameter until a critical size of 1–2 μm (dependent on the stress intensity factor) above which it becomes independent of particle size.

Unfortunately, no experimental data on crack propagation in tablet film coatings exist. However, data on the incidence of edge cracking on tablet film coatings containing different pigments (Rowe, 1982b; Gibson et al., 1988) do show that there are minimal differences between the incidence of the defect for formulations containing red iron oxide (mean particle size 0.5–1 μm) compared with those containing aluminium lake pigments (mean particle size 1–2 μm) at equivalent volume fraction.

Conclusion

Despite the fact that the program contains many simplifications of a very complex phenomenon, the agreement with experiment both qualitatively and quantitatively is extremely good. The program is very easy to use, enabling many simulations to be performed with minimum effort. In this work only a single population of inclusions with a constant standard deviation of mean diameter (10%) has been evaluated. It is

possible to run the program with two populations of differing sizes, each with different standard deviations and this will be the subject of a future publication.

Acknowledgments

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