Proceedings of the 6th International Conference on Recrystallization and Grain Growth (ReX&GG 2016) *Edited by: Elizabeth A. Holm, Susan Farjami, Priyadarshan Manohar, Gregory S. Rohrer, Anthony D. Rollett, David Srolovitz, and Hasso Weiland*
TMS (The Minerals, Metals & Materials Society), 2016

Theoretical Basis of the Aboav-Weaire Equation and Topological Consequences

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Keywords: Aboav equation, Topological correlation, Statistical theory

Introduction

The Aboav-Weaire (AW) equation [1,2] describes a topological correlation, in any 2-d grain microstructure, between the number of sides of a given grain *m* and the average number of sides of the first neighbours of the grain *m*.
The origin of this equation is essentially of an empirical nature, although a similar equation has already been theoretically derived within the framework of the Statistical Theory of Grain Growth (STGG) by the present Author [3]. The essential difference between the two equations, which are actually of the same formal nature, is the mathematical dimension in which the equation is described. The original Aboav-Weaire equation has been formulated (empirically derived and formally guessed) in the mathematical space in which the independent variable is *m*, namely the number of grain sides (or corners), whereas the STGG derives the equation adopting the grain size *R* (equivalent radius) as the independent variable and the number of grain corners as the dependent one (in this case giving the so-called Special Linear Relationship in the space (*m*, *R*) [3]). On the other hand, the topological meaning and statistical nature of the equations was clearly explained as a direct consequence of the STGG in terms of the higher probability of the small grains having larger grains as neighbours and just the opposite for the larger grains.
A formulation of the basic equations of the STGG in the dimension *m* is reported here, with the derivation of the various statistical and topological quantities having *m* as the independent variable, allowing to derive the original AW expression and to discuss other kinetics and topological consequences of interest for the general Theory.

Theoretical Derivation of STGG in the dimension *m*

Here, the grains belonging to the statistical classes with the same number of sides (*m* or *n*) will be identified as *m* and *n*. Moreover $\varphi(m)$ or φ_m represents the frequency distribution function of the grains having a different number of sides *m* (in fact an histogram, being *m* an integer variable). Moreover, being φ_m the frequency of *m* grains in the system, then $\sum_m \varphi_m = 1$ for *m*=1,…N_s, where N_s is the number of grain classes *m* describing the grain system. η_{mn} will represent the maximum number of *n*-sided grains which can stay in contact with the grain of *m* sides. Therefore by using the Von Neumann- Mullins approach, one can write:

$$\eta_{mn}\left(\beta_{mn} - \alpha_{mn}\right) = 2\pi \quad (1)$$

with β_{mn} equal to the inclination angle at any vertex of the microstructure made of a grain *m* surrounded by grains *n*, while α_{mn} represents the inclination angle along each grain side *mn* (see also [4]). From the STGG one can also write:

$$\varphi_m w_{mn} \eta_{mn} = \varphi_n w_{nm} \eta_{nm} \tag{2}$$

expressing the obvious symmetry conditions that the number of contacts among grains having a number of sides *m* and the grains of sides *n* are the same as those of *n* grains with *m* grains in the given microstructure (w_{mn} here being the probability that *n* grains are in contact with an *m* grain, namely $\sum_n w_{mn} = 1$ for the sum on *n* for *n* = 1 to N_s, total number of classes).

The probability factor w_{mn} (compare with w_{ij} in [5]) has the dependency also on *m*, meaning that in principle grains of different classes *m* have the possibility of being in contact with grain *n*, the different probability depending specifically on *m* and not only on *n*. This means that a correlation which is of a physical nature, besides that of geometrical origin involved in conditions (1) and (2) , can also be considered.

STGG has been typically formulated in the grain size dimension [3], identifying grain size by means of the Perimeter *P* or an equivalent radius *R* from the grain Area. Dividing the grain size dimension into classes, these are identified by the indices i,j,… with i and j ranging in the interval 1 to N_c = total number of classes considered in the system. So the probability of contact between an *i* grain and a *j* grain is there written w_{ij}. Based on the existence of simple geometrical rules, an equivalent expression to eq. (2) for size classes, and symmetry requirements, one can suppose (or theoretically derive by induction) that $w_{ij} = w_j$ (randomness condition) so that the probability of contacts with any *j* grains will be the same for each *i* grain, i.e. the contact probability function will be independent of *i*. Here, in the dimension of grain sides (corners), we will introduce the randomness condition using an explicit averaging procedure which will clarify the difference between the two dimensions (size and sides).

Hence, the fact that the probability of contacts with any grain of *n* sides will be the same for each *m* grain, i.e. that w_{mn} will be independent of *m*, is derived by averaging on the *m* grains the total inclination curvature given by eq. (1) and deriving the expression w_n as follows [6]:

$$w_n = \frac{\sum_m \varphi_m w_{mn} \eta_{mn} (\beta_{mn} - \alpha_{mn})}{2\pi \sum_m \varphi_m} \tag{3}$$

it being also $\alpha_{mn} = -\alpha_{nm}$ and using eq. (2), one obtains : $w_n = \left(\frac{1}{2\pi}\right) \sum_m \varphi_n w_{nm} \eta_{nm} (\beta_{mn} + \alpha_{nm})$

and because, in case of uniform grain boundary energy (absence of texture), $\beta_{mn} = \beta_{nm} = \frac{\pi}{3}$ is valid, it follows from eq. (1):

$$w_n = \left(\frac{1}{2\pi}\right) \sum_m \varphi_n w_{nm} \eta_{nm} \left(\frac{\pi}{3} + \alpha_{nm}\right) = \left(\frac{1}{2\pi}\right) \sum_m \varphi_n w_{nm} \eta_{nm} \left(\frac{2\pi}{3} - \frac{2\pi}{\eta_{nm}}\right) = \varphi_n \frac{(n-3)}{3} \tag{5}$$

or more generally (keeping in mind that $\sum_n \varphi_n n = 6$ is also valid)

$$w_n = \frac{\varphi_n (n-3)}{\sum_n \varphi_n (n-3)} \tag{6}$$

with $0 < w_n < 1$ as a normal probability function for which $\sum_n w_n = 1$. The probability function w_n described by eq. (6) includes the topological information that in each corner only three grains meet and that the average value of sides for such a microstructure is 6. Therefore the smallest *n* grains (near to disappearing from the microstructure, because of a 120° angle at the vertices) tend to have 3 sides and zero probability of contacts with the other grains. Obviously, another reason for an *n*-class grain to have zero probability of contact with other grains is its having zero frequency (φ_n=0) in the system. A grain having a number of sides (*n*=6) has a probability $w_n = \varphi_n$ which depends only on the frequency of the grain *n* in the system (a grain of *n*=6 sides, equal to the average value, is topologically "neutral" in the microstructure).

The expression for the parameter η_{nm}

By using the previous result for w_n, Eq. (2) will change as follows:

$$\varphi_m w_n \eta_{mn} = \varphi_n w_m \eta_{nm} \tag{7}$$

which together with eq. (1), by substituting η_{nm}, will lead to :

$$2\pi\varphi_m w_n = \varphi_n w_m \eta_{nm}\left(\beta_{mn} + \alpha_{nm}\right) = \varphi_n w_m \eta_{nm}\left(\beta_{mn} + \beta_{nm} - \left(\beta_{nm} - \alpha_{nm}\right)\right)$$

Which, in case of uniform grain boundary energy, becomes:

$$2\pi\varphi_m w_n = \varphi_n w_m \eta_{nm}\frac{2\pi}{3} - 2\pi\varphi_n w_m$$

$$\varphi_n w_m \frac{\eta_{nm}}{3} = \varphi_m w_n + \varphi_n w_m$$ and by substituting eq. (5) (or (6)) for w_n and w_m it ensues that:

$$\eta_{nm} = \frac{\left[3(n-3)+3(m-3)\right]}{(m-3)} = 3\left[1 + \frac{(n-3)}{(m-3)}\right] \tag{8}$$

Correspondingly, the following symmetrical expression can also be derived:

$$\eta_{mn} = 3\left[1 + \frac{(m-3)}{(n-3)}\right] \tag{9}$$

From eq. (9) it is possible to see that the maximum number η_{mn} of *n* grains around an *m* grain depends only on the number of sides *n* and the number of sides of grain *m*. In particular a grain *m* having only 3 sides, *m* = 3, can accommodate only three grains : $\eta_{mn} = 3$ from eq. (9).

Obviously grains with an equal number of sides *n* and *m* will lead to $\eta_{mn} = 6$.

In the case of the size dimension (described by the continuous function P_i, perimeter of grain *i*), the number n_{ij} of adaptable grains *j* around a grain *i* is in general a non-integer number. This is a direct consequence of the calculation of n_{ij} as a ratio between the total available contact perimeter of a grain *i* and the given contact perimeter of a single boundary of a grain *j*, which therefore is a continuous function. Here, due to the same concept, η_{mn} can be fractional (although *n* and *m* are always integer numbers), where the calculation based on grain perimeter is substituted with that based on grain excess curvature from eq. (1). As will be shown later, the average procedure on the total microstructure (grain classes), typical of the STGG, will mitigate

this apparent problem, which is more mathematical than physical, leading to more simple and robust equations where only statistical parameters are involved (which are the only ones easily obtainable by experimental measurements).

Kinetics equation in the dimension of sides (corners) *m* of the grains

Starting from eq. (1) one can also write:

$$\eta_{mn} = \frac{2\pi}{\left(\beta_{mn} - \alpha_{mn}\right)} = \frac{2\pi}{\theta_{mn}} \tag{10}$$

Using a similar statistical procedure as in [5], one can write for the kinetics of the area A_m of grain *m*:

$\frac{dA_m}{dt} = \sum_n w_n \eta_{mn} \left[\frac{dA_m}{dt}\right]_n$ where $\left[\frac{dA_m}{dt}\right]_n$ is the contribution to the kinetics of grain *m* caused by contact with a grain *n*. It is proportional to the curvature $\alpha_{mn} = \beta_{mn} - \frac{2\pi}{\eta_{mn}}$ of the boundary *mn*, such that, using the expressions for w_n and η_{mn} , with $\beta_{mn} = \frac{\pi}{3}$ and M being the grain boundary diffusivity, one obtains:

$\frac{dA_m}{dt} = \sum_n \varphi_n \frac{(n-3)}{3} \cdot 3\left[1 + \frac{(m-3)}{(n-3)}\right] \cdot M\left(\frac{\pi}{3} - \frac{2\pi}{\eta_{mn}}\right)$ which by simple calculation leads to :

$$\frac{dA_m}{dt} = \frac{\pi}{3} M(m-6) \tag{11}$$

which is exactly the Von Neumann equation expressing the kinetics of growth for a grain having a number of sides *m*. Therefore the classical Von Neumann equation is a consequence of averaging the kinetics on single boundaries *mn* with statistical parameters obtained by averaging on the boundary excess curvature instead of on grain perimeter and under the randomness assumption of eq. (3).

In case the average procedure of the STGG were carried out in the dimension of grain perimeters, the equivalent result of eq. (10) is:

$\frac{dA_i}{dt} = \frac{\pi}{3} M\left(\overline{n_i} - 6\right)$ where $\overline{n_i}$ is equal to the average number of sides for *i* size (perimeter/equivalent radius) grains. The expression of $\overline{n_i}$ as a function of Perimeter or equivalent radius represents the so-called SLR (Special Linear Relationship) [3].

Exact derivation of the Aboav-Weaire equation by STGG

The equations to remember here are those for w_n and η_{mn} (eq. (5), (9)) and eq. (10) for the angle of contact. In the AW equation, the *n* grains to be considered are the first neighbours of grain *m* for which the average number of contacts must be calculated (on the generic surrounding grains *p*) considering the concomitant contact of the *n* grains with *m* grains (see the scheme in fig. 1, which has been plotted using simple circles just to point out such angular interference [3]).

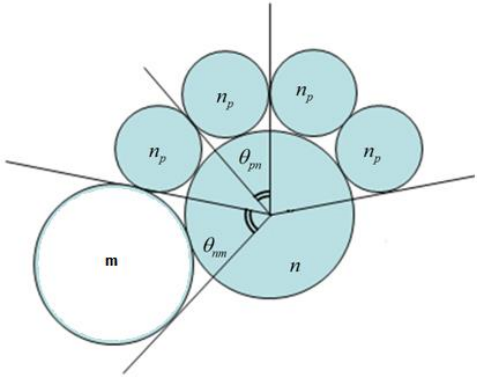

Fig. 1. Diagram showing the influence of the inner grain *m* on the number of sides of the first neighbours.
The number of sides of the *n* grains, forming the set of grains, first neighbours of the *m* grain, is influenced by the presence of the *m* grain itself, through the engagement of the boundary *mn* under the covering angle θ_{nm}. This "contact" angle will no longer be available for contact with the *p* grains.

Grain *n* has limited surrounding space to be filled with the various *p* grains due to the contact with the *m* grain (θ_{nm} measures the engaged angle). Therefore, in evaluating the number of contacts (sides) of the *n* grains (first neighbours of the grain *m*), the following condition must be taken into account: the average procedure for finding the number of possible contacts of grain *n* must be extended to the possible surrounding *p* grains, under the condition that a fixed contact with grain *m* is always present, thus limiting the available contact space for *p* grains.
Because the maximum number of surrounding grains *p* around grain *n* is given by:

$$\eta_{np} = 3\left[1+\frac{(n-3)}{(p-3)}\right] = \frac{2\pi}{\theta_{np}} = \frac{2\pi}{\left(\frac{\pi}{3}-\alpha_{np}\right)}$$

In case of concomitant contact with grain *m*, one can write for the number of contacts *np*:

$$\eta_{np}' = 1+\frac{(2\pi-\theta_{nm})}{\frac{\pi}{3}-\alpha_{np}} = 1+\frac{\left(2\pi-(\frac{\pi}{3}-\alpha_{nm})\right)}{\frac{\pi}{3}-\alpha_{np}} = 1+\frac{2\pi}{\frac{\pi}{3}-\alpha_{np}} - \frac{(\frac{2\pi}{\frac{\pi}{3}-\alpha_{np}})}{(\frac{2\pi}{\frac{\pi}{3}-\alpha_{nm}})} = 1+\eta_{np}-\frac{\eta_{np}}{\eta_{nm}}$$

Again one can see that the maximum number of *p* grains around an *n* grain is "influenced" by *m* through η_{nm}. Now, averaging the contacts for the *n* grain, in the presence of a neighbour *m,* we shall have:

$$\eta_{n,m} = \sum_p w_p \eta_{np}' = 1+n-\frac{n}{\eta_{nm}} = 1+n-n\frac{(m-3)}{(3(n+m-6))} \quad \text{it being that} \quad \sum_p w_p \eta_{np} = n,$$

therefore the average number of sides of the first neighbours of grain *m* will be:

$$\overline{m}^s = \frac{\sum_n w_n \eta_{mn} \eta_{n,m}}{\sum_n w_n \eta_{mn}} = \frac{\sum_n w_n \eta_{mn} \eta_{n,m}}{m} = \frac{1}{m}\sum_n \varphi_n \frac{(n-3)}{3}\cdot 3\left[1+\frac{(m-3)}{(n-3)}\right]\cdot\left[1+n-n\frac{(m-3)}{3(n+m-6)}\right] =$$

$$= \frac{1}{3m}\sum_n \varphi_n (n+m-6)\cdot\frac{\left[3(1+n)(n+m-6)-n(m-3)\right]}{(n+m-6)} = \frac{1}{m}\left(-30+5m+\overline{n^2}\right)$$

and it being that $\overline{n^2} = \left(\overline{n}\right)^2 + \sigma_n^2 = 36 + \sigma_n^2$ one obtains:

$$\overline{m}^s = 5 + \frac{(6 + \sigma_m^2)}{m} \tag{12}$$

which is exactly the AW equation [2] ($\sigma_n^2 = \sigma_m^2$ is the variance of the same grain population).

Topological Consequence of the AW equation

The original eq. (12), in its dependency on *m* and the presence of the constant 5, has been empirically derived. The presence of an additional quantity in the numerator of eq. (12), (6 + σ_m^2), was identified, by measurements carried out by Aboav [1], as a quantity that was systematically greater than 6, and such deviation was subsequently "guessed" by Weaire [2] as something that could be of the order of the "variance parameter" σ_m^2, though no theoretical explanation for this was given. Here instead it is shown that eq. (12) is not only an empirical relationship but is topologically explainable and has its theoretical foundation in the STGG.
In this framework, this equation is exactly derivable using the dimension of grain sides *m*. The phenomenological result that the first neighbours of large grains tend to have a smaller average number of sides, while the exact opposite happens for small grains, is strictly linked to how, in the structural pattern consisting of "inner grain and first neighbours", the size of the inner grain influences the available space for the contact grains of the first neighbours and to the expression of the probability function (eq.(6)). The latter does not imply any physical correlation among grains (randomness) in addition to the one imposed by the geometrical constraints of eq.(10).
In a previous paper [3], a similar equation to eq. (12) was derived using the dimension of grain size (*R* equivalent radius). In the space (*m*, *R*), the special linear relationship is valid between grains in the class of size R_i and the corresponding dependent variable $\overline{m_i}$, i.e.

$$\overline{m_i} = 3 + 3\frac{R_i}{\overline{R}} = 3 + 3\, r_i \tag{13}$$

with $\overline{R}$ being the mean radius of grain population and $r_i = \frac{R_i}{\overline{R}}$ the normalized grain size. The corresponding derivable Aboav equation in the r_i dimension has the following expression:

$$\overline{m_i^s} = 5 + \frac{(6 + \sigma_{\overline{m}_i}^2)}{\overline{m}_i} \tag{14}$$

as can be seen, the difference with eq. (12) is only of a formal nature, $\overline{m}_i$ being the average number of sides of the grains of size R_i (or r_i). The topological meaning is the same of that of eq. (12) because the derivation is carried out by the same conceptual approach as in the grain size dimension and using the SLR. Therefore, the validity of the WA equation (14) holds true under the same conditions as the SLR [3]. Moreover, eq. (14) shows that it is always $\overline{m}_i^s > 5$, and, for smaller grains ($\overline{m}_i \to 3$) , it follows that $\overline{m}_i^s = 7 + \frac{\sigma_{\overline{m}_i}^2}{3}$ which is significantly larger than 6.

Furthermore, in this R_i dimension it is also possible to formulate, on the same theoretical basis, an equation which gives the mean radius $\overline{R}_i^s$ of the surrounding grains (first neighbours) as a function of the grain size of the inner grain, namely an equivalent AW equation for the dimension of grain size, obtaining the following expression [3]:

$$\bar{r}_i^s = (1 + \frac{k_i^2}{1 + r_i}) \tag{15}$$

with $r_i = \frac{R_i}{\overline{R}}$, $\bar{r}_i^s = \frac{\overline{R}_i^s}{\overline{R}}$, $k_i = \frac{\sigma_i}{\bar{r}} = \sigma_i$ the variation coefficient ($\sigma_i^2 = \sum_i \varphi_i (r_i - \bar{r})^2 = \overline{r_i^2} - 1$).

Comparison between eq. (12) and (14) and with eq. (15) immediately shows that for large grains *i*, i.e. with a large number of sides, the average number of sides of the surrounding grains is 5 while the corresponding average grain size is $\overline{R}$, as could normally be expected and has been verified in experiments [3,6]. This represents a first source of scattering in the correlation between grain size and the corresponding number of sides of the grains due to topological reasons (inherent). In fact, while the average number of sides of first neighbours is strongly conditioned by contact with the inner grain, the average grain size of the first neighbours does not have such a topological constraint [6]. Therefore, the number of corners can deviate from the correlation expected for the average behaviour expressed by the SLR due to specific topological restrictions.

Moreover, since the SLR (eq. (13)) describes the average behaviour of any grain in the system, one can also use this equation for grains of size $\bar{r}_i^s$ to calculate the corresponding expected average value of $\overline{m}_i$ as $\overline{m}_i^s = 3 + 3\,\bar{r}_i^s$. Substituting here eq. (15), one has $\overline{m}_i^s = 3 + 3\,(1 + \frac{k_i}{1 + r_i})$ which for $r_i \to 0$ gives $\overline{m}_i^s = 6 + 3\,k_i^2 = 6 + 3\,\sigma_i^2$ and it being that $\sigma_{\overline{m}_i}^2 = 9\,\sigma_i^2$ it follows that:

$\overline{m}_i^s = 6 + \frac{\sigma_{\overline{m}_i}^2}{3}$; to be compared with $\overline{m}_i^s = 7 + \frac{\sigma_{\overline{m}_i}^2}{3}$ obtained from eq. (14) for $\overline{m}_i \to 3$ (which corresponds to $r_i \to 0$ in eq. (15)). Therefore the limit results for the deviations are:

$$\overline{m}_i^s(\,\overline{m}_i \to 3\,) - \overline{m}_i^s(r_i = 0) = 7 - 6 = 1 \tag{16}$$

$$\overline{m}_i^s(\,\overline{m}_i \to \infty) - \overline{m}_i^s(r_i \to \infty) = 5 - 6 = -1 \tag{17}$$

These results show that due to the topological constraints put in the grain patterns, made of "inner grain and first neighbours", the grain system is forced to locally deviate from the general topological rule identified by the SLR. This is responsible for a scattering around the SLR, enclosed in a band around the SLR, amounting to a maximum width of plus or minus 1 grain side with respect to the SLR average values (see eq. (16), (17) and fig.2). In other words, observing the typical scattering of data in 2D microstructures (m, R space), one can see that more than approximately 70% of total deviation from the SLR is due to simultaneous geometrical and topological constraints of the WA and equation (15) and only the rest is dependent on normal statistical scattering (measurement error, lack of homogeneity conditions, 2D-cut of 3D microstructure,…).

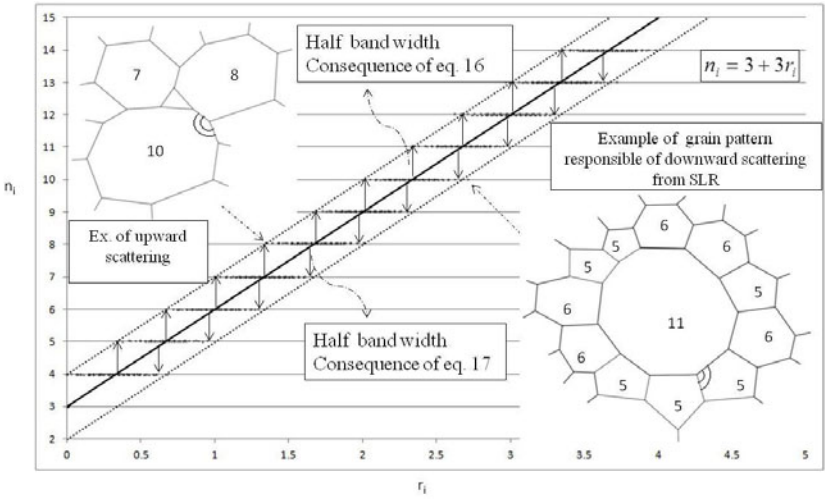

Fig. 2. Scattering around the SLR induced by the concurrent validity of AW and eq. (15).

On the other hand, such a topological "constraint", of which the AW equations (12) and (14) and eq. (15) are the consequences, has considerable effects also on the kinetics of grain growth in 2D, which in fact is controlled by the number of corners of the grains (eq. (11)). Indeed, the possibility that grains of a given size can have a scattering of +/- 1 in the number of sides can influence the kinetics of grain size (Area) change, introducing a "noise" around the growth kinetics of the grains following the SLR law "on average". This effect is expected to influence more significantly the shape of the grain size distribution (in particular that of steady state condition in the case of normal grain growth) rather than the averaged microstructure parameters such as: grain mean radius/perimeter/area, average boundary length, volume fraction of specific grain texture components, etc., which are essentially "integral" parameters.
Finally, the above treatment can be extended to the 3D case by deriving equivalent equations (AW type and eq.(15)) that, although formally more complex, lead to the same type of topological consequences, namely an intrinsic scattering around the average behaviour of the relationship between the number of grain faces and grain size [7].

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