A Simple Iterative Method for Analysing Experimental Tracer Diffusivities in Concentrated Binary Alloys

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ABSTRACT

In this paper, we present an iterative method for analysing experimental tracer diffusivities in binary disordered alloys using the self-consistent theory of Moleko *et al*. (1989). The method can be used for any binary system that can be approximated by a random alloy. Some aspects of the convergence of the proposed numerical procedure are discussed. We determine tracer correlation factors and ratios of exchange frequencies from ratios of measured tracer diffusivities for the following systems: Cu-Pt, Zr-U, Mo-W, Ag-Au, Fe-Ni, Co-Ni, Cr-Ni, Cu-Ni, Fe-Pd, Pb-Tl, Nb-Ti, Mo-Ni and Fe-Mn.

1. Introduction

In order to obtain the tracer correlation factors f_A and f_B in a concentrated binary alloy AB there are essentially two methods available. The first method is based on the measurement of the isotope effect for one or both components. For the usual vacancy mechanism this measurement gives the tracer correlation factor multiplied by the kinetic factor ΔK, see, for example, [1]. The ΔK factor has the limits $0.0 \leq \Delta K \leq 1.0$ but its actual value in a given situation is rather poorly known, though in close packed metals it is thought to be about 0.9 [2]. Measurement of the isotope effect is a difficult and time consuming experiment compared with measurements of the tracer diffusivities themselves [3]. Moreover, frequently, appropriate isotopes are simply not available. The second method centres on the use of a model to convert values of both tracer diffusivities D_{A*} and D_{B*} to the individual tracer correlation factors. It has long been known that the random alloy is a useful one for describing tracer diffusion kinetics in concentrated disordered alloys [4,5]. In the usual form of this model, the atomic components are randomly mixed and isolated vacancies at vanishingly small concentrations provide the vehicles of motion. There is no binding of the vacancy to either component. The atom-vacancy exchange frequencies are denoted by w_i. Taken at face value, the model might imply that the w_i are independent of local environment. We cannot expect real alloys to behave quite in this way of course because the environment obviously varies from site to site and with composition. This was recognized by Manning [4,5], who considered the frequencies in the random alloy model to be averaged quantities. Manning derived the following closed-form expressions for the tracer correlation factors [4,5]:

$$f_A^{Man} = f_0 \left(1 - \frac{2c_B(D_{A*} - D_{B*})}{M_0(c_a D_{A*} + c_B D_{B*})} \right) \qquad (1)$$

$$f_B^{Man} = f_0 \left(1 + \frac{2c_A(D_{A*} - D_{B*})}{M_0(c_a D_{A*} + c_B D_{B*})} \right) \qquad (2)$$

where c_i is the atomic fraction and $M_0 = 2f_0/(1-f_0)$ where f_0 is the geometric tracer correlation factor for the lattice, e.g. $f_0 = 0.78146\ldots$ for the f.c.c. lattice [6]. Recent extensive Monte Carlo calculations have shown that Manning's diffusion kinetics formalism for the random alloy is not particularly accurate [7-11]. Another formalism addressing diffusion kinetics in the random alloy model is that due to Holdsworth and Elliott [12]. The results of this formalism are in rather better agreement with the results of Monte Carlo calculations. Finally, there is the self-consistent formalism of Moleko, Allnatt and Allnatt (MAA) [13], which provides almost exact agreement with the Monte Carlo results. Even when the ratio of the atom-vacancy exchange frequencies is within a factor of two or three, the MAA formalism is to be preferred over those of Manning [4,5] and Holdsworth and Elliott [12]. The MAA expressions for the tracer correlation factors are as follows:

$$f_A^{MAA} = \frac{P_A + \sqrt{P_A^2 + 2M_0 H w_B G_A}}{G_A} \tag{3}$$

$$f_B^{MAA} = \frac{P_B + \sqrt{P_B^2 + 2M_0 H w_A G_B}}{G_B} \tag{4}$$

where

$$P_A = H(H - M_0 w_B)$$
$$\tag{5}$$
$$P_B = H(H - M_0 w_A)$$

and

$$G_A = 2(H + 2(c_A w_B + c_B w_A))(H + 2w_A)$$
$$\tag{6}$$
$$G_B = 2(H + 2(c_A w_B + c_B w_A))(H + 2w_B)$$

$$H = h + \sqrt{h^2 + 2M_0 w_A w_B} \tag{7}$$

$$\text{with} \quad h = \frac{c_A w_A + c_B w_B}{1 - f_0} - w_A - w_B \tag{8}$$

It is immediately clear that the MAA approach for obtaining the tracer correlation factors is rather cumbersome to use, requiring some numerical effort to solve the system of equations (Eqns 3-8). In the present paper, we propose a simple iterative method for employing the MAA approach. We demonstrate its use with application to a number of systems where experimental tracer diffusivity data exist for both components.

2. Theory

In order to implement the MAA theory for analysing tracer diffusivities in disordered alloys we make use of the following iterative procedure. In the binary random alloy both components 'see' the vacancy equally. We then have that $D_{A*}/D_{B*} = f_A w_A/f_B w_B$. Let us assume that we have values for the tracer diffusivities in hand. We can now build iterations using the following recurrence relation:

$$x_i = \frac{D_{A*}}{D_{B*}} \frac{f_B^{i-1}}{f_A^{i-1}} \tag{11}$$

where f_A^{i-1} and f_B^{i-1} can be calculated using the following (scaled) values: $w_A = x_{i-1}$, $w_B = 1$ with values of c_A and c_B (=$1-c_A$) given. For convenience, one of the exchange frequencies is put equal to unity. These values are to be substituted into Eqns 3-8 to obtain the required values of f_A^{i-1} and f_B^{i-1}. For convenience, it is useful to write the initial value for $x_0 = D_{A*}/D_{B*}$.

Now let us discuss convergence matters associated with the procedure. Let us assume that $w_A < w_B$. (The following analysis can be easily repeated for $w_A > w_B$.)

Statement 1. For all compositions of the random alloy, if $w_A < w_B$ then $f_A > f_0$ whereas $f_B < f_0$ [7]. Therefore, $f_B/f_A < 1.0$. Moreover, if we know that $w_A^i/w_B^i < w_A^{i-1}/w_B^{i-1}$ then $f_B^i/f_A^i < f_B^{i-1}/f_A^{i-1}$.

Now, let us denote $x_0 = D_{A*}/D_B < 1.0$. We notice now that all $x_i = w_A^i/w_B^i$ are constructed in such a way that, for example, (Statement 1):

$$x_1 = x_0 \, f_B^0/f_A^0 < x_0$$

then (using again Statement 1):

$$x_2 = x_0 \, f_B^1/f_A^1 < x_0 \, f_B^0/f_A^0 = x_1 < x_0$$

and repeating this procedure we will have for any n:

$$x_n < x_{n-1} < \ldots < x_1 < x_0. \tag{12}$$

Now, if we denote $x = w_A/w_B$ (exact ratio of the atom-vacancy exchange frequencies), then we soon have that $x_0 > x$ ($x_0 \neq 1.0$). Now, assuming that $x_i > x$ we are going to prove by induction that $x_{i+1} > x$. The proof is in noting that for $x_i > x$ we will have that (Statement 1):

$$\frac{f_B^i}{f_A^i} > \frac{f_B}{f_A} \tag{13}$$

and therefore:

$$x_{i+1} - x = \frac{D_{A*}}{D_{B*}} \frac{f_B^i}{f_A^i} - \frac{D_{A*}}{D_{B*}} \frac{f_B}{f_A} = \frac{D_{A*}}{D_{B*}} \left(\frac{f_B^i}{f_A^i} - \frac{f_B}{f_A} \right) > 0 \tag{14}$$

which leads to:

$$w_A/w_B < x_i \quad \text{for any i.} \tag{15}$$

Relations 12 and 15 guarantee convergence of the proposed iterative procedure.

Let us consider compositions where $c_A < f_0$, i.e. above the percolation threshold or 'forbidden region' [4,5]. Then to obtain a result accurate, say, to the fifth figure, the number of required iterations will be about 10. On the other hand, if $c_A \geq f_0$, i.e. compositions are below the percolation threshold, then the number of the iterations will be proportional to the value w_B/w_A. If we can restrict ourselves *a priori* to $w_B/w_A \approx 100$ then the upper limit (with respect to composition) for the number of iterations is found to be about 500. However, there is another issue to be considered for this composition range. The ratio of the tracer diffusivities must be less than the limits provided in the theories of MAA and Manning (these limits are numerically quite similar and are of the order $1/(1-f_0)$, see also [9].

Experimental ratios of tracer diffusivities greater than these limits must then be interpreted with the formal inclusion of solute-vacancy binding. Below the percolation threshold, the result of any numerical calculations (even using the simpler Manning approach) will be extremely sensitive to any sources of error in the tracer diffusivities. Fig. 1 shows the typical convergence of x_i $(= w_A^i/w_B^i)$ as a function of the number of iterations at various compositions. Here $x = 0.01$, $f_0 = 0.7272$ (b.c.c. lattice). We always used the ratio of tracer diffusion coefficients as the starting value for x_0 (points corresponding to unity on the abscissa).
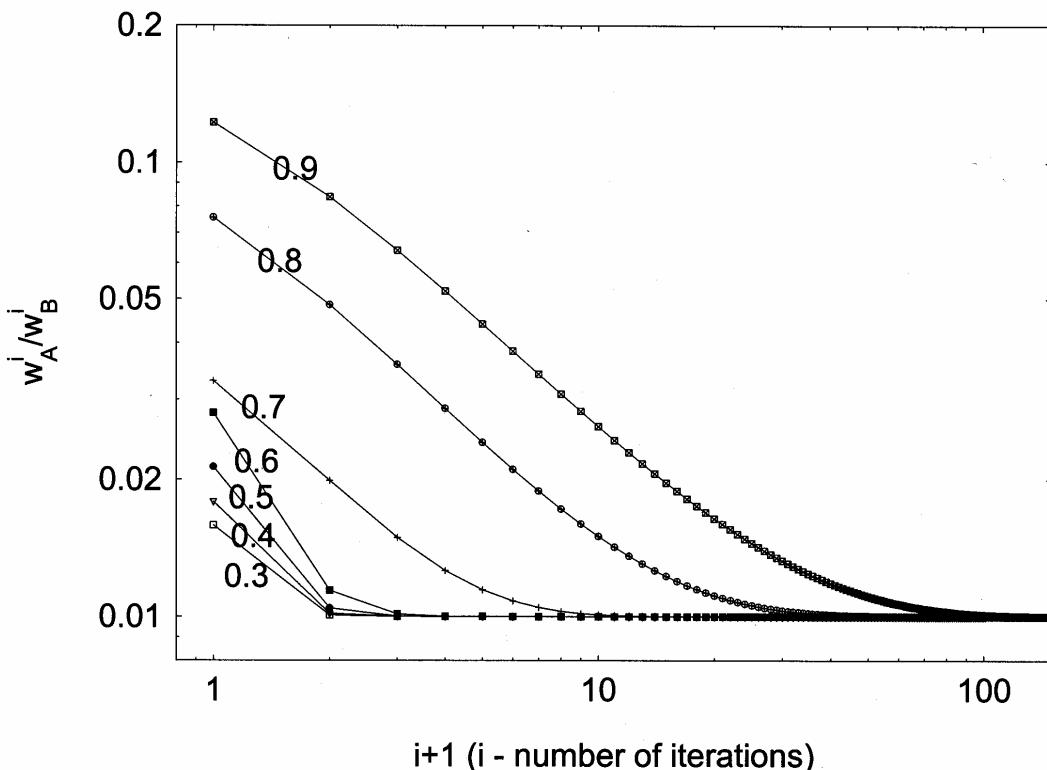

Figure 1. Ratios of w_A^i/w_B^i as functions of the number of iterations i. Curves correspond to different compositions c_A. The curves $c_A = 0.8$ and 0.9 are below the percolation threshold. The exact value of $w_A/w_B = 0.01$.

3. Applications

As examples for using the above iterative procedure, we have determined tracer correlation factors and ratios of exchange frequencies from ratios of measured tracer diffusivities for a number of binary alloys. The results are presented in the tables for the following systems: Cu-Pt (Table 1), Zr-U (Table 2a,b), Mo-W (Table 3), Ag-Au (Table 4), Fe-Ni (Table 5), Co-Ni (Table 6a,b), Cr-Ni (Table 7), Cu-Ni (Table 8), Fe-Pd (Table 9), Pb-Tl (Table 10a,b), Nb-Ti (Table 11), Mo-Ni (Table 12), Fe-Mn (Table 13).

Table 1, Tracer correlation analysis for the system Cu-Pt, T=1300K [14].

c_{Pt}	f_{Pt}	f_{Cu}	D_{Pt*}/D_{Cu*}	w_{Pt}/w_{Cu}
0.098	0.9670	0.7431	0.0910	0.0700
0.246	0.9383	0.6817	0.1285	0.0934
0.494	0.9029	0.4989	0.0907	0.0501
0.745	0.8456	0.3094	0.1612	0.0590

Table 2a, Tracer correlation analysis for the system Zr-U, T=1175K and 1249K [15].

	T=1175K				T=1249K			
c_{Zr}	f_{Zr}	f_U	D_{Zr*}/D_{U*}	w_{Zr}/w_U	f_{Zr}	f_U	D_{Zr*}/D_{U*}	w_{Zr}/w_U
0.410	0.3768	0.8405	5.5355	12.3489	0.5406	0.8087	2.5655	3.8379
0.610	0.8534	0.1261	0.0016	0.0002	0.8534	0.1258	0.0014	0.0002

Table 2b, Tracer correlation analysis for the system Zr-U, T=1323K [15].

c_{Zr}	f_{Zr}	f_U	D_{Zr*}/D_{U*}	w_{Zr}/w_U
0.410	0.7779	0.6854	0.7036	0.6200
0.610	0.8534	0.1255	0.0011	0.0002

Table 3, Tracer correlation analysis for the system Mo-W, T=2100K and 2600K [16]. (

	T=2100K				T=2600K			
c_W	f_W	f_{Mo}	D_{W*}/D_{Mo*}	w_W/w_{Mo}	f_W	f_{Mo}	D_{W*}/D_{Mo*}	w_W/w_{Mo}
0.250	0.3531	0.7914	3.4944	7.8324	0.7024	0.7350	1.1232	1.1754
0.350	0.6736	0.7523	1.3198	1.4740	0.7548	0.7111	0.8486	0.7996
0.500	0.8537	0.4822	0.2178	0.1230	0.8325	0.5531	0.3388	0.2251
0.650	0.8156	0.4114	0.2652	0.1338	0.7556	0.6652	0.7280	0.6409

Table 4. Tracer correlation analysis for the system Ag-Au, T=1000K and 1220K [17].

	T=1000K				T=1220K			
c_{Au}	f_{Au}	f_{Ag}	D_{Ag*}/D_{Au*}	w_{Ag}/w_{Au}	f_{Au}	f_{Ag}	D_{Ag*}/D_{Au*}	w_{Ag}/w_{Au}
0.080	0.9321	0.7604	0.2691	0.2195	0.9011	0.7666	0.4173	0.3550
0.170	0.9168	0.7370	0.2921	0.2348	0.8879	0.7505	0.4384	0.3706
0.350	0.8902	0.6856	0.3148	0.2424	0.8645	0.7180	0.4654	0.3865
0.500	0.8612	0.6548	0.3744	0.2846	0.8448	0.6919	0.4897	0.4011
0.660	0.8381	0.6021	0.3829	0.2750	0.8213	0.6753	0.5413	0.4450
0.830	0.8059	0.6025	0.4796	0.3586	0.7983	0.6753	0.6181	0.5229
0.940	0.7900	0.5834	0.5010	0.3700	0.7873	0.6624	0.6286	0.5288

Table 5, Tracer correlation analysis for the system Fe-Ni, T+1260K and 1560K [18].

	T=1260K				T=1560K			
c_{Ni}	f_{Ni}	f_{Fe}	D_{Ni*}/D_{Fe*}	w_{Ni}/w_{Fe}	f_{Ni}	f_{Fe}	D_{Ni*}/D_{Fe*}	w_{Ni}/w_{Fe}
0.149	0.8469	0.7675	0.6565	0.5950	0.8366	0.7701	0.7097	0.6533
0.297	0.7998	0.7732	0.8828	0.8535	0.8424	0.7493	0.6242	0.5552
0.453	0.8079	0.7568	0.7887	0.7387	0.8037	0.7611	0.8210	0.7775
0.605	0.8041	0.7414	0.7562	0.6972	0.8231	0.6971	0.5765	0.4882
0.700	0.7879	0.7656	0.9044	0.8788	0.7837	0.7760	0.9655	0.9560
0.753	0.7906	0.7511	0.8412	0.7992	0.7939	0.7386	0.7880	0.7331
0.798	0.7852	0.7661	0.9184	0.8961	0.7881	0.7531	0.8581	0.8201

Table 6a, Tracer correlation analysis for the system Co-Ni, T=1370K and 1470K [19].

	T=1370K				T=1470K			
c_{Ni}	f_{Ni}	f_{Co}	D_{Ni*}/D_{Co*}	w_{Ni}/w_{Co}	f_{Ni}	f_{Co}	D_{Ni*}/D_{Co*}	w_{Ni}/w_{Co}
0.037	0.8751	0.7768	0.5580	0.4953	0.8527	0.7782	0.6632	0.6052
0.071	0.8672	0.7124	0.4565	0.3749	0.8705	0.7727	0.5677	0.5039
0.300	0.8564	0.6428	0.3773	0.2832	0.8733	0.7677	0.5415	0.4760
0.103	0.9019	0.7615	0.4021	0.3395	0.8565	0.7387	0.5406	0.4663
0.493	0.8238	0.5814	0.3942	0.2782	0.8412	0.7017	0.5208	0.4344
0.697	0.7842	0.5254	0.4369	0.2927	0.8161	0.6734	0.5554	0.4583
0.737	0.7847	0.4540	0.3701	0.2141	0.8141	0.6533	0.5277	0.4235

Table 6b, Tracer correlation analysis for the system Co-Ni, T=1570K [19].

c_{Ni}	f_{Ni}	f_{Co}	D_{Ni*}/D_{Co*}	w_{Ni}/w_{Co}
0.037	0.8067	0.7804	0.8803	0.8516
0.071	0.8199	0.7782	0.8118	0.7705
0.103	0.8047	0.7786	0.8821	0.8535
0.300	0.7852	0.7798	0.9753	0.9685
0.493	0.8006	0.7611	0.8345	0.7934
0.697	0.7939	0.7499	0.8236	0.7780
0.737	0.7847	0.7721	0.9445	0.9293

Table 7, Tracer correlation analysis for the system Cr-Ni, T=1225K and 1465K [20, 21].

	T=1225K				T=1465K			
c_{Ni}	f_{Ni}	f_{Cr}	D_{Ni*}/D_{Cr*}	w_{Ni}/w_{Cr}	f_{Ni}	f_{Cr}	D_{Ni*}/D_{Cr*}	w_{Ni}/w_{Cr}
0.523	0.8524	0.6627	0.4154	0.3229	0.8462	0.6782	0.4614	0.3698
0.616	0.8479	0.6027	0.3531	0.2510	0.8415	0.6307	0.4073	0.3053
0.656	0.8280	0.6524	0.4814	0.3793	0.8263	0.6593	0.4978	0.3972
0.706	0.8224	0.6363	0.4752	0.3676	0.8255	0.6199	0.4435	0.3330
0.764	0.8078	0.6670	0.5714	0.4718	0.8094	0.6577	0.5502	0.4471
0.780	0.8185	0.5662	0.4017	0.2779	0.8219	0.5317	0.3594	0.2325
0.857	0.7955	0.6729	0.6225	0.5265	0.7981	0.6456	0.5656	0.4575

Table 8, Tracer correlation analysis for the system Cu-Ni, T=1380K [22, 14].

c_{Ni}	f_{Ni}	f_{Cu}	D_{Ni*}/D_{Cu*}	w_{Ni}/w_{Cu}
0.215	0.9182	0.7188	0.2571	0.2013
0.546	0.8001	0.7569	0.8219	0.7776
0.870	0.8094	0.4094	0.3040	0.1538

Table 9, Tracer correlation analysis for the system Fe-Pd, T=1373K and 1523K [23].

	T=1373K				T=1523K			
c_{Pd}	f_{Pd}	f_{Fe}	D_{Pd*}/D_{Fe*}	w_{Pd}/w_{Fe}	f_{Pd}	f_{Fe}	D_{Pd*}/D_{Fe*}	w_{Pd}/w_{Fe}
0.100	0.8231	0.7763	0.7907	0.7458	0.8205	0.7767	0.8035	0.7606
0.200	0.8468	0.7614	0.6391	0.5746	0.8264	0.7686	0.7493	0.6969
0.300	0.8494	0.7441	0.5824	0.5102	0.8269	0.7587	0.7156	0.6566
0.400	0.8360	0.7355	0.6146	0.5407	0.8281	0.7437	0.6675	0.5995
0.500	0.8281	0.7222	0.6130	0.5346	0.8260	0.7256	0.6291	0.5526
0.550	0.8239	0.7154	0.6133	0.5326	0.8204	0.7222	0.6419	0.5651
0.600	0.8218	0.7029	0.5935	0.5077	0.8155	0.7182	0.6510	0.5734
0.700	0.8073	0.7065	0.6532	0.5716	0.8024	0.7235	0.7130	0.6429
0.800	0.7937	0.7246	0.7480	0.6829	0.7929	0.7291	0.7639	0.7024
0.900	0.7865	0.7303	0.7912	0.7347	0.7885	0.7067	0.7204	0.6457

Table 10a, Tracer correlation analysis for the system Pb-Tl, T=480K and 580K [24].

	T=480K				T=530K			
c_{Tl}	f_{Tl}	f_{Pb}	D_{Tl*}/D_{Pb*}	w_{Tl}/w_{Pb}	f_{Tl}	f_{Pb}	D_{Tl*}/D_{Pb*}	w_{Tl}/w_{Pb}
0.100	0.5603	0.7972	2.1987	3.1284	0.6169	0.7944	1.8766	2.4166
0.200	0.6080	0.8104	2.1112	2.8139	0.6441	0.8061	1.8582	2.3258
0.340	0.6292	0.8326	2.3066	3.0521	0.6586	0.8258	2.0063	2.5155
0.500	0.6423	0.8659	2.9244	3.9426	0.6685	0.8557	2.4205	3.0981
0.620	0.6560	0.8970	3.9825	5.4459	0.6739	0.8873	3.2345	4.2586
0.740	0.6825	0.9305	6.2772	8.5585	0.6947	0.9204	4.6823	6.2039
0.760	0.6921	0.9329	6.1798	8.3308	0.7019	0.9238	4.7535	6.2565

Table 10b, Tracer correlation analysis for the system: Pb-Tl, T=580K [24].

c_{Tl}	f_{Tl}	f_{Pb}	D_{Tl*}/D_{Pb*}	w_{Tl}/w_{Pb}
0.100	0.6905	0.7897	1.4742	1.6860
0.200	0.6938	0.7990	1.5297	1.7618
0.340	0.7007	0.8140	1.6221	1.8845
0.500	0.7080	0.8364	1.8145	2.1435
0.620	0.7031	0.8681	2.3558	2.9088
0.740	0.7171	0.8981	2.9955	3.7517
0.760	0.7196	0.9042	3.1869	4.0045

Table 11, Tracer correlation analysis for the system: Nb-Ti, T=1280K and 1573K [25].

	T=1280K				T=1560K			
c_{Nb}	f_{Nb}	f_{Ti}	D_{Nb*}/D_{Ti*}	w_{Nb}/w_{Ti}	f_{Nb}	f_{Ti}	D_{Nb*}/D_{Ti*}	w_{Nb}/w_{Ti}
0.643	0.6330	0.8411	2.4356	3.2366	0.6613	0.8158	1.8736	2.3112
0.804	0.6840	0.8533	2.2113	2.7587	0.7124	0.7805	1.3116	1.4369
0.946	0.7174	0.8562	1.9760	2.3582	0.7209	0.8179	1.5342	1.7404

Table 12, Tracer correlation analysis for the system: Mo-Ni, T=1373K and 1573K [26].

	T=1373K				T=1573K			
c_{Ni}	f_{Ni}	f_{Mo}	D_{Ni*}/D_{Mo*}	w_{Ni}/w_{Mo}	f_{Ni}	f_{Mo}	D_{Ni*}/D_{Mo*}	w_{Ni}/w_{Mo}
0.770	0.7681	0.8210	1.2970	1.3864	0.7753	0.8011	1.1296	1.1671
0.800	0.7691	0.8249	1.3198	1.4154	0.7736	0.8105	1.1954	1.2525
0.820	0.7664	0.8393	1.4575	1.5962	0.7711	0.8232	1.2955	1.3830
0.840	0.7758	0.8090	1.1740	1.2242	0.7814	0.7818	1.0017	1.0022
0.920	0.7795	0.8033	1.1209	1.1551	0.7778	0.8204	1.2375	1.3054

Table 13, Tracer correlation analysis for the system: FeMn, T=1270K and 1510K [27].

	T=1270K				T=1510K			
c_{Mn}	f_{Mn}	f_{Fe}	D_{Mn*}/D_{Fe*}	w_{Mn}/w_{Fe}	f_{Mn}	f_{Fe}	D_{Mn*}/D_{Fe*}	w_{Mn}/w_{Fe}
0.070	0.5085	0.7938	2.4185	3.7757	0.6215	0.7902	1.8121	2.3041
0.104	0.6126	0.7952	1.9067	2.4750	0.6703	0.7916	1.5861	1.8730
0.182	0.6084	0.8073	2.0690	2.7454	0.7213	0.7929	1.3475	1.4813
0.255	0.6854	0.8069	1.6408	1.9316	0.7889	0.7789	0.9545	0.9423

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