How Dr. Malcom M. Cross may have tackled the development of "An apparent viscosity function for shear thickening fluids"

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Abstract

A new apparent viscosity function for shear thickening fluids has been recently published [1], which is able to cover the three characteristic regions typically exhibited by these materials (thinning/thickening/thinning). The proposed function was shown to provide an excellent fitting to several independent data sets used. However, although its formulation has the Cross model as a starting point, the K_i-parameter time constants were enforced to be negative, which is counter-intuitive. In the light of the original work of Cross [2], the present short communication introduces small changes to the viscosity function developed in [1] to make the fitting parameters more natural. The original microstructural ideas of Cross are also considered in order to provide additional intuition and interpretation. Finally, the fitting

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values of the model parameters for all of the curves considered in the original manuscript are included here so that results can be reproduced or used for numerical simulations.

Keywords: Generalized Newtonian Fluid, viscosity function, shear thickening fluids, rheology.

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1. Introduction

Galindo-Rosales et al. [1] have recently proposed Eq. 1 as a mathematical expression capable of accurately describing the rate dependent steady shear viscosity $\eta(\dot{\gamma})$ of shear thickening fluids (STF's). The viscosity function is a branched equation based on the Cross model. It considers the three regimes typically present in the general form of a STF viscosity curve, capturing the shear thinning ($\dot{\gamma} \leq \dot{\gamma}_c$), shear thickening ($\dot{\gamma}_c < \dot{\gamma} \leq \dot{\gamma}_{max}$) and second shear thinning regime ($\dot{\gamma}_{max} < \dot{\gamma}$). An advantage of the proposed constitutive relationship is that it provides a function with a continuous derivative, which is useful for computational codes to solve complex flows using a Generalized Newtonian Fluid (GNF) model.

$$\eta\left(\dot{\gamma}\right) = \begin{cases} \eta_I\left(\dot{\gamma}\right) = \eta_c + \frac{\eta_0 - \eta_c}{1+\left[K_I\left(\frac{\dot{\gamma}^2}{\dot{\gamma}-\dot{\gamma}_c}\right)\right]^{n_I}} & \text{for } \dot{\gamma} \leq \dot{\gamma}_c \\ \\ \eta_{II}\left(\dot{\gamma}\right) = \eta_{max} + \frac{\eta_c - \eta_{max}}{1+\left[K_{II}\left(\frac{\dot{\gamma}-\dot{\gamma}_c}{\dot{\gamma}-\dot{\gamma}_{max}}\right)\dot{\gamma}\right]^{n_{II}}} & \text{for } \dot{\gamma}_c < \dot{\gamma} \leq \dot{\gamma}_{max} \\ \\ \eta_{III}\left(\dot{\gamma}\right) = \frac{\eta_{max}}{1+\left[K_{III}\left(\dot{\gamma}-\dot{\gamma}_{max}\right)\right]^{n_{III}}} & \text{for } \dot{\gamma}_{max} < \dot{\gamma} \end{cases} \tag{1}$$

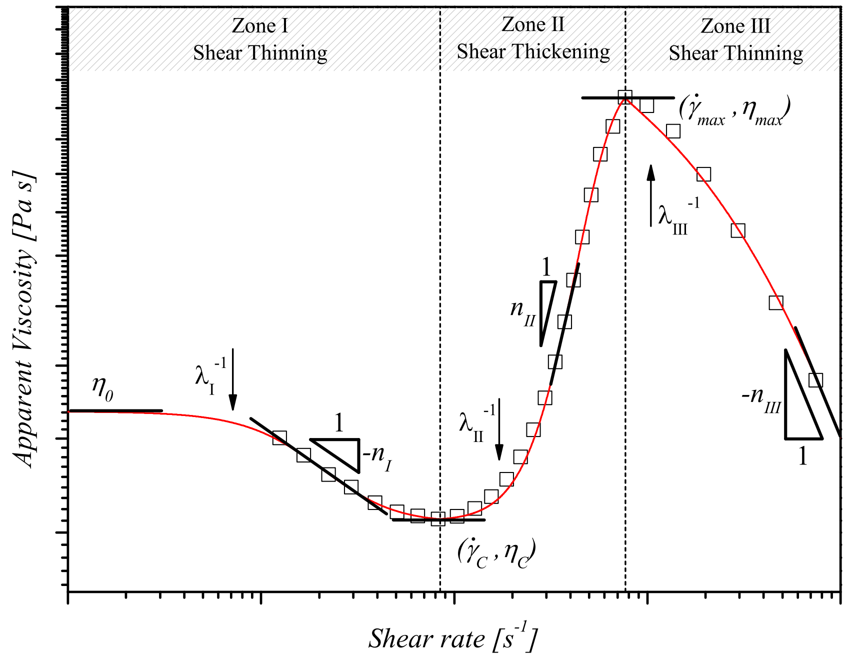

Figure 1: The three typical regions (thinning/thickening/thinning) of the viscosity curve of a Shear Thickening Fluid in a log-log plot (see Appendix A for fitting parameters).

As a major drawback, Eq. 1 possesses a high number of independent constants. Since the relationship is purely empirical, the parameters are determined by fitting available experimental data. The 11 independent parameters are connected to the shape of the viscosity curve (Fig. 1).

2. Modification to enforce positive time constants

In Eq. 1 confusion will result if all parameters are assumed to be positive. Specifically, positive K_i values would produce complex numbers for the bracketed terms in branches I and II, when taken to an arbitrary power n_i. This is simply due to the choice of sign in the denominator of these bracketed terms resulting in negative values, i.e. the $(\dot{\gamma} - \dot{\gamma}_c)$ term in Region I, and the $(\dot{\gamma} - \dot{\gamma}_{max})$ term in Region II. In each case the term is negative, and taking an arbitrary power n_i of a negative number results in a complex number. (To be precise, the special case when n_i is a rational power $n_i = a/b$ written in lowest terms with b odd will produce a real number, but we wish to allow arbitrary values for n_i.) In the original publication [1] the K_i time constant parameters were negative ($i = I, II$), the bracketed terms were therefore positive, and complex numbers were avoided.

Negative K_i time constants are counter-intuitive, and potentially confusing. To avoid the peculiarity of negative time constants, we suggest a simple sign modification for branches I and II, which will allow for positive time constants $\lambda_i = -K_i$, while $\lambda_{III} = K_{III}$ in Region III. We change the notation to λ_i to avoid confusion with the previous negative constants K_i in Eq. 1 and the original publication [1]. The following improved expression should replace Eq. 1.

$$\eta(\dot{\gamma}) = \begin{cases} \eta_I(\dot{\gamma}) = \eta_c + \frac{\eta_0 - \eta_c}{1 + \left[\lambda_I \left(\frac{\dot{\gamma}^2}{\dot{\gamma}_c - \dot{\gamma}}\right)\right]^{n_I}} & \text{for } \dot{\gamma} \le \dot{\gamma}_c \\ \eta_{II}(\dot{\gamma}) = \eta_{max} + \frac{\eta_c - \eta_{max}}{1 + \left[\lambda_{II}\left(\frac{\dot{\gamma} - \dot{\gamma}_c}{\dot{\gamma}_{max} - \dot{\gamma}}\right)\dot{\gamma}\right]^{n_{II}}} & \text{for } \dot{\gamma}_c < \dot{\gamma} \le \dot{\gamma}_{max} \\ \eta_{III}(\dot{\gamma}) = \frac{\eta_{max}}{1 + [\lambda_{III}(\dot{\gamma} - \dot{\gamma}_{max})]^{n_{III}}} & \text{for } \dot{\gamma}_{max} < \dot{\gamma} \end{cases} \tag{2}$$

The model parameters are typically determined from simple shear flow data [1]. In general, and for simulations, Eq. 2 can be used in a Generalized Newtonian Fluid constitutive model with viscosity coefficient $\eta(\dot{\gamma})$. Then the shear rate magnitude $\dot{\gamma}$ is the scalar invariant of the strain rate tensor $\dot{\gamma}_{ij} = \frac{\partial}{\partial x_i} v_j + \frac{\partial}{\partial x_j} v_i$, such that $\dot{\gamma} = \sqrt{\frac{1}{2} \sum_i \sum_j \dot{\gamma}_{ij} \dot{\gamma}_{ji}}$.

In Eq. 2, the λ_i parameters are positive characteristic time constants. The manuscript of Galindo-Rosales et al. [1] did not contain any discussion about the physical meaning of the time constant parameters. Since the Cross model is taken as the starting point here, we revisit the microstructural considerations of Cross' original work [2] and then discuss their possible interpretation in the case of shear thickening fluids.

3. The Microstructural Basis of the Cross Model

3.1. Background: The original Cross model

Dr. Malcom M. Cross proposed a steady shear viscosity function intended for shear thinning systems [2]. It was originally presented in the form

$$\eta = \eta_\infty + \left(\frac{\eta_0 - \eta_\infty}{1 + \alpha \dot{\gamma}^m}\right), \tag{3}$$

where η_0 is the limiting viscosity at zero rate of shear, η_∞ is the limiting viscosity at infinite rate of shear, α and m are two independent constants, and $\dot{\gamma}$ is the rate of shear. Note that α is not raised to the power n with Cross' original notation, in contrast with the form of Eq. 1 and Eq. 2.

Cross was pursuing a flow equation relating the steady shear viscosity and the shear rate, and proposed the following requirements (in Cross' words):

1. It should give an accurate fit of experimental data over a wide range of shear rates.
2. It should involve a minimum number of independent constants.
3. The appropriate constants should be readily evaluated.
4. The constants should have a real physical significance.

We recall that Cross developed Eq. 3 from microstructural ideas, specifically principles of reaction kinetics for the formation and rupture of linkages between particles. His assumptions are summarized in the following list:

1. Shear-thinning is associated with the transient formation and rupture of structural linkages, associated with the flocculation behavior of particles.
2. Under shear conditions there is an average group size of particles dependent on the magnitude of the applied shear rate. These groups are assumed to take the form of random linked chains.
3. There is a total number of single particles (P) per unit volume, linked or otherwise.
4. There is an average number of links per chain (L) at shear rate ($\dot{\gamma}$). At zero rate of shear, this is a fixed number of links per chain (L_0), while at

very high rates of shear the system becomes completely deflocculated and L tends to zero.

5. Linkages break down under shear with a first-order rate constant $k_0 + k_1\dot{\gamma}^m$, where $\dot{\gamma}^m$ was assumed to be an even function.
6. At the same time, linkages are being formed with a first-order rate constant k_2, e.g. as a result of Brownian movement.

Thus, with all these assumptions, the time dependence of the average number of linkages L is described by the following rate equation which is a competition between creation and destruction,

$$\frac{dL}{dt} = k_2 P - [k_0 + k_1\dot{\gamma}^m]\, L. \tag{4}$$

The equilibrium is attained when $\frac{dL}{dt} = 0$, in which case the steady state result is obtained, $\frac{L}{L_0} = \frac{1}{1+\alpha\dot{\gamma}^m}$.

Finally, Cross assumed that the viscosity is directly proportional to the number of links per chain: $\eta = \eta_\infty + BL$, where B is a constant. Then, since $\eta = \eta_0$ when $L = L_0$, one can derive Cross' viscosity function, Eq. 3, which satisfies all the requirements listed above. The parameter $\alpha = \frac{k_1}{k_0}$ is then interpreted as a ratio of rate constants for shear-dependent structural breakdown (k_1) to zero-shear structural breakdown (k_0). Is is also apparent, as Cross noted, that $\alpha^{-1/m}$ represents a characteristic shear rate at which the viscosity of the system is the mean of the two limiting values η_0 and η_∞.

3.2. Constant α versus λ

Nowadays rheologists do not make use of the Cross model in the original form given by Eq.3, but in the following form:

$$\eta = \eta_\infty + \frac{\eta_0 - \eta_\infty}{1 + (\lambda\dot{\gamma})^n}, \qquad (5)$$

Here n is an arbitrary dimensionless power-law exponent, similar to m in Eq. 3, but with no restriction on $\dot{\gamma}^n$ being an even function (the restriction is unnecessary since $\dot{\gamma}$ is always the positive scalar magnitude of the shear rate tensor $\dot{\gamma}_{ij}$). The parameter λ is positive and interpreted as a time constant of the fluid; it therefore has the dimensions of time [3]. It is analogous to the time constant in the Carreau-Yasuda equation [4, 5].

The constant α from the original Cross model (Eq. 3) and the constant λ (Eq. 5) are linked by the following expression:

$$\alpha = \lambda^n. \qquad (6)$$

4. Extending Cross' Ideas to Shear Thickening Fluids

Considering the four requirements that Cross considered for the formulation of a flow equation relating the viscosity and shear rate, it is obvious that the proposed viscosity function for STF's, Eq. 2, is not entirely satisfactory in all four respects. In particular, it does not accomplish criterion #4, according to which the parameters involved in the viscosity function should have a real physical significance. To fill this current deficiency, here we discuss the possible extension of Cross' microstructural ideas to shear thickening fluids.

We must notice that STF's are normally formed by non-flocculated suspensions where particle affinity is inhibited and, consequently, under rest conditions particles do not form a 3D network, although they can form aggregates [6–9]. The presence of these aggregates and their disaggregation under shear would justify the light shear thinning behavior at low shear rates (below the critical point) [10], and this is qualitatively consistent with Cross' microstructural consideration. An additional shear-induced microstructural change is known to occur in the shear-thinning Region I. Shear thinning may originate from particles re-arranging from a disordered state to a layered state [3]. Either or both of these microstructural transitions cause shear-thinning (Region I). Shear thinning occurring at high shear rates, beyond $\dot{\gamma}_{max}$ (Region III), has been explained in three different ways [11–14]: (1) the specimen fractures owing to the high viscosity; (2) hydrocluster aggregates are broken into successively smaller units as the shear rate increases; (3) at very high stresses, particles cease to behave like rigid bodies, and start to deform elastically, which alters their rheology and, subsequently, the effect can be understood as a manifestation of the finite elasticity of the particles.

If these shear thinning regions (I and III) of the viscosity curve are directly interpreted under the assumptions made by Cross for shear-thinning behavior, then the α_i-parameters could be obtained for both branches of the viscosity curve ($\alpha_i = \frac{k_{1_i}}{k_{0_i}}$, for $i = I, III$). In this way it would be possible to give physical meaning to the time constant, defined as a ratio between the shear and Brownian contributions to the link rupture rate constant: $\lambda_i = \alpha_i^{1/n_i} = \left(\frac{k_{1_i}}{k_{0_i}}\right)^{1/n_i}$, for $i = I, III$.

The shear thickening behavior, Region II, requires more careful consideration. According to the works of Wagner and collaborators [15–19], shear thickening behavior is accepted to occur by the formation of "hydroclusters", jammed clusters resulting from hydrodynamic lubrication forces between particles [20]. In a very simplistic way, it can be thought that the increase of viscosity (Region II) starts from a critical point ($\dot{\gamma}_c$) in which there is a "weak equilibrium" set by the balance of interparticle and hydrodynamic forces. The Péclet number is a dimensionless number that relates the particle advection rate to the particle diffusion rate ($Pe = \frac{\dot{\gamma}a^2}{D_0}$). When $Pe \approx 1$, as anticipated by Brady [21], the crossover from shear thinning to shear thickening occurs [22]. An increase in the shear contribution ($Pe > 1$) leads particles to form hydroclusters, due to hydrodynamic interactions, overcoming the existing repulsive forces between them. Then, in this region of the viscosity curve, we can consider that shear forces help to construct the hydroclusters, while Brownian forces (plus repulsive interparticle forces) try to separate the particles. These cluster formations increase the hydrodynamic stress in the suspension, thus marking the region of shear thickening behavior. The hydroclustered state, however, does not lead to permanent or irreversible particle aggregation [18, 20].

Considering the hydrocluster formation and the assumptions described by Cross in his original work, α_{II} could be derived as $\alpha_{II} = \frac{k_{1_{II}}}{k_{0_{II}}}$. Thus, the time constant λ_{II} would be defined as a ratio between the shear and Brownian contributions to the link *formation* rate constant: $\lambda_{II} = \alpha_{II}^{1/n_{II}} = \left(\frac{k_{1_{II}}}{k_{0_{II}}}\right)^{1/n_{II}}$.

Considering the work by Cross for the formulation of the flow equation for shear thinning materials and applying all the assumptions he made to the formulation of a viscosity equation for STF's, given by Eq. 2, each time constant λ_i ($i = I, II, III$) has a positive value and could be understood as the ratio between the shear and Brownian contributions to the kinetics of linkage rupture or formation, depending on the region in the viscosity curve. Nevertheless, we caution that the specific kinetic assumptions of Cross have no reason to be precisely applied to shear thickening systems. In particular, the assumed power law dependence on the shear-dependent rupture/formation is not rooted in a specific theory. Further work is obviously required in this sense, to have a flow curve which naturally arises from the microstructural considerations specific to shear thickening fluids, which have been summarized by Wagner and Brady [14].

5. Conclusion

In spite of the good performance of Eq. 1, as clearly demonstrated in [1], the K_i-parameters are not properly introduced if we consider the Cross model as a starting point for its formulation. Inspired by the original work of Cross [2], in this communication we have developed a deeper analysis of this equation, leading to a slight modification of the apparent viscosity function proposed in [1] that is more intuitive than the original one, having positive time constants λ_i. In addition, the parameters appearing in Eq. 2 can be given a direct microstructural interpretation, with some insight gained by considering Cross' original arguments for his model. We recommend the use of Eq. 2 instead of Eq. 1 developed in the original manuscript [1]. The Appendix includes all model parameters used for the data in the original manuscript [1], using the parameters defined in Eq. 2.

Appendix A. Fitting parameters

Table A.1: Fitting parameters of the viscosity curves shown in the work of Galindo-Rosales et al. [1] considering the parameters defined by Eq. 2. SI units are used, i.e. viscosities η in [Pa s], time constants λ_i in [s], shear rates $\dot{\gamma}$ are [s^{-1}], and power law exponents n_i are dimensionless [-]. Note that Fig. 1 in both this manuscript and in Galindo-Rosales et al. [1] is the same as Fig.2.b listed in the table.

Curve	η_0	λ_I	n_I	η_c	$\dot{\gamma}_c$	λ_{II}	n_{II}	η_{max}	$\dot{\gamma}_{max}$	λ_{III}	n
Fig2.a	0.832 ± 0.001	1.80 ± 0.02	0.83 ± 0.01	0.599 ± 0.003	10.91 ± 0.05	1.3 ± 0.1	2.1 ± 0.2	2.17 ± 0.07	129 ± 4	$(18 \pm 1)\,10^{-4}$	1.01
Fig2.b	0.869 ± 0.002	1.97 ± 0.02	0.984 ± 0.007	0.625 ± 0.002	8.26 ± 0.03	0.019 ± 0.001	1.2 ± 0.1	2.27 ± 0.05	77 ± 2	$(21 \pm 1)\,10^{-4}$	0.90
Fig2.c	2.7 ± 0.1	8.67 ± 0.09	0.69 ± 0.05	2.44 ± 0.07	1.67 ± 0.05	0.13 ± 0.01	1.01 ± 0.05	6.26 ± 0.05	56.2 ± 0.4	$(39 \pm 1)\,10^{-4}$	1.55
$Fig3_{\phi=0.40}$	136.1 ± 0.9	$(60 \pm 4)\,10^{3}$	0.61 ± 0.03	0.916 ± 0.003	3.336 ± 0.003	0.85 ± 0.06	1.3 ± 0.1	1.79 ± 0.02	69.18 ± 0.09	$(37 \pm 1)\,10^{-5}$	0.71
$Fig3_{\phi=0.45}$	256.3 ± 0.3	$(28 \pm 2)\,10^{3}$	0.62 ± 0.05	1.76 ± 0.08	1.43 ± 0.04	1.30 ± 0.04	1.6 ± 0.1	10.02 ± 0.06	12.5 ± 0.9	$(14 \pm 1)\,10^{-4}$	0.65
$Fig3_{\phi=0.48}$	137.9 ± 0.2	$(40 \pm 3)\,10^{3}$	0.55 ± 0.05	2.59 ± 0.09	0.96 ± 0.06	1.3 ± 0.1	2.49 ± 0.06	42.33 ± 0.03	3.7 ± 0.4	$(23 \pm 1)\,10^{-3}$	0.80
$Fig3_{\phi=0.49}$	385 ± 7	$(28 \pm 2)\,10^{3}$	0.61 ± 0.01	10.99 ± 0.04	0.113 ± 0.005	9.5 ± 0.7	2.9 ± 0.1	292.6 ± 0.2	0.86 ± 0.08	$(92 \pm 2)\,10^{-3}$	0.826
Fig4	42 ± 2	$(159 \pm 3)\,10^{4}$	0.42 ± 0.07	0.28 ± 0.01	5.5 ± 0.1	0.040 ± 0.002	2.00.2	19.26 ± 0.03	72.5 ± 0.4	–	
$Fig5_{2.51g/ml}$	–	–	–	182.2 ± 0.6	1.26 ± 0.02	0.17 ± 0.01	1.1 ± 0.1	305 ± 4	26.3 ± 0.7	$(150 \pm 9)\,10^{-4}$	1.23
$Fig5_{2.54g/ml}$	–	–	–	294 ± 2	0.53 ± 0.08	0.11 ± 0.05	1.08 ± 0.03	626 ± 7	11.1 ± 0.5	$(263 \pm 9)\,10^{-4}$	1.25
$Fig5_{2.56g/ml}$	–	–	–	472 ± 8	0.11 ± 0.01	0.32 ± 0.02	0.94 ± 0.06	1777 ± 12	5.93 ± 0.03	$(85 \pm 2)\,10^{-3}$	1.21
$Fig6_{20\%}$	3.72 ± 0.02	0.06 ± 0.02	1.17 ± 0.01	2.56 ± 0.05	221 ± 4	$(45 \pm 8)\,10^{-5}$	1.63 ± 0.09	22 ± 1	902 ± 7	$(214 \pm 7)\,10^{-5}$	1.88
$Fig6_{30\%}$	5.79 ± 0.09	0.034 ± 0.009	1.10 ± 0.02	3.75 ± 0.07	121 ± 1	$(10 \pm 4)\,10^{-4}$	2.06 ± 0.04	34.35 ± 0.03	581 ± 2	$(41 \pm 3)\,10^{-5}$	0.85
$Fig6_{40\%}$	10.71 ± 0.08	0.036 ± 0.001	0.88 ± 0.02	5.98 ± 0.06	139 ± 7	$(11 \pm 2)\,10^{-4}$	2.25 ± 0.07	34.2 ± 0.4	540 ± 3	$(91 \pm 2)\,10^{-5}$	1.13
$Fig6_{0kAm}$	16.3 ± 0.7	23.9 ± 0.5	0.50 ± 0.01	1.5 ± 0.3	219 ± 2	$(41 \pm 2)\,10^{-5}$	1.35 ± 0.02	25 ± 2	1305 ± 5	$(124 \pm 4)\,10^{-5}$	1.31
$Fig6_{100kAm}$	163 ± 2	84 ± 7	0.48 ± 0.02	3.86 ± 0.04	326 ± 3	$(24 \pm 2)\,10^{-4}$	1.84 ± 0.07	31 ± 1	734 ± 11	$(70 \pm 2)\,10^{-5}$	1.23
$Fig6_{200kAm}$	860 ± 20	333 ± 11	$(577 \pm 8)\,10^{-3}$	10 ± 1	217 ± 9	$(29 \pm 2)\,10^{-4}$	1.2 ± 0.1	33 ± 2	599 ± 7	$(84 \pm 1)\,10^{-5}$	1.26

Table A.2: Coefficients of determination R_i^2 of the fitting procedures corresponding to the different regions of the viscosity curves shown in the work of Galindo-Rosales et al. [1] and considering Eq. 2.

Curve	R_I^2	R_{II}^2	R_{II}^2
Fig2.a	0.99955	0.93028	0.94033
Fig2.b	0.99805	0.91596	0.94537
Fig2.c	0.99535	0.92451	0.98128
Fig3$_{\phi=0.40}$	0.99601	0.96565	0.97358
Fig3$_{\phi=0.45}$	0.99288	0.92662	0.98288
Fig3$_{\phi=0.48}$	0.99201	0.96135	0.98958
Fig3$_{\phi=0.49}$	0.99704	0.91399	0.99419
Fig4	0.99438	0.97823	–
Fig5$_{2.51g/ml}$	–	0.92489	0.98883
Fig5$_{2.54g/ml}$	–	0.91435	0.99049
Fig5$_{2.56g/ml}$	–	0.91242	0.99745
Fig6$_{20\%}$	0.97593	0.96683	0.99937
Fig6$_{30\%}$	0.94194	0.96399	0.98736
Fig6$_{40\%}$	0.93042	0.90716	0.99643
Fig6$_{0kAm}$	0.98411	0.90415	0.99812
Fig6$_{100kAm}$	0.90732	0.99996	0.99748
Fig6$_{200kAm}$	0.98995	0.90892	0.95284

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