

## The Effect of Particle Size Distributions on the Entropy in Magnetic Fine Particles System

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### Abstract:

The magnetic entropy change curves are calculated numerically as a function of magnetic field, absolute temperature, magnetic interactions, and the effect of anisotropy are taken into account. In these calculations we utilize different types of distribution functions of particles size, i.e., uniform distribution (UD or DD), Log-normal distribution (LD), Gaussian distribution (GD), and Rectangular distribution (RD or CD). In all calculations we assume a system of Fe<sub>3</sub>O<sub>4</sub> fine particles system. In this paper we present the effect of particles size distributions on the magnetic entropy change in magnetic fine particles system were calculated numerically as a function of mean volume of particles ( $\bar{V}$ ), the standard deviation ( $\sigma$ ), the magnetic field ( $H$ ), and the absolute temperature ( $T$ ). For each curve the results were compared with the corresponding results obtained using the classical theory of superparamagnetism. Four distribution functions were taken Uniform distribution (UD or DD), Constant distribution (CD), Gaussian distribution (GD) and Log-Normal distribution (LD).

### Index Terms:

Effects of Magnetic interactions, mean volume, standard deviation, average anisotropy constant and particle size distributions.

### I. INTRODUCTION:

Decreasing the width of the distribution actually increasing the percentage of the optimized particles The  $\Delta S(T)$  curves can be understood with the help of the sum rule.

$$\text{i.e. } \int_0^M \Delta S dT = -M_0 H$$

This sum rule implies that for materials with the same saturation magnetic moment  $M_0$ , those with the high entropy change at a given temperature will have low entropy changes at there temperatures,

and those materials which do not have a large entropy change at any particular temperature can undergo a moderate entropy change over a broader temperature range. Thus for the distribution (50-60 Å) which show large  $\Delta S$  for  $T < 90\text{k}$ , but show smaller  $\Delta S$  for  $T > 90\text{k}$ . With increasing the applied magnetic field, the number of particles align with the field is increased, thus the change in magnetic entropy  $\Delta S$  is increased accordingly. The change in magnetic entropy increases with increasing the volume exhibiting a maximum. With decreasing  $\sigma$  not only the position of the maximum  $\Delta S$  increases, but also the shape of the maximum becomes narrower. Due to the number of optimized volume is increased. Increasing the energy barrier  $\Delta S$  is increased, or equivalently the effective volume is increased and hence the particles are easily aligned with the field, thus the  $\Delta S$  is increased.

### a. The effect of particle size distributions:

It is clear from figures 3.1-3.4, that with the exception of rectangular distribution (which is not suitable for ultra fine particle systems), the magnetic entropy change is not affected by the kind of distribution assumed. This may be due to the fact that there is a certain particle size, which contributes significantly to the magnetic entropy change. For the case of rectangular distribution, large portion of particles lies in the size range which corresponds to the maximum entropy change, therefore, using the RD corresponds to  $\Delta S$  greater than their corresponding using GD or LD.

### b. The effect of standard deviation:

The effect of  $\sigma$  (or the width of the distributions) as shown in Fig.3.2 and Fig.3.5. This behavior can be explained as follows, keeping the average volume constant and varying the width (10-100 Å), (30-80 Å), (50-60 Å), the magnetic entropy change increases with decreasing  $\sigma$  (or decreasing the width of the distribution). Decreasing the width of the distribution actually increasing the percentage of the optimized particles, i.e., particles which shows

maximum entropy change. Note that

the optimized volume is  $\bar{D} \approx 48 \text{ \AA}$  ( $x=3.5$ , corresponds to  $\bar{D} \approx 48 \text{ \AA}$ ). For (10-100  $\text{\AA}$ ) the particle sizes show greater deviation from optimum, but for (30-80  $\text{\AA}$ ) show less deviation from optimum, whereas for (50-60  $\text{\AA}$ ) show lesser deviation from optimum.

### c. The effect of temperature T:

The  $\Delta S(T)$  curves can be understood with the help of the sum rule. i.e.

$$\int_0^M \Delta S dT = -M_0 H$$

this sum rule implies that for materials with the same saturation magnetic moment, those with the high entropy change at a given temperature will have low entropy changes at other temperatures, and those materials which do not have a large entropy change at any particular temperature can undergo a moderate entropy change over a broader temperature range. Thus for the distribution (50-60  $\text{\AA}$ ) which show large  $\Delta S$  for  $T < 90\text{k}$ , but show smaller  $\Delta S$  for  $T > 90\text{k}$ .

### d. The effect of applied field H:

With increasing the applied magnetic field, the number of particles align with the field is increased, thus the change in magnetic entropy  $\Delta S$  is increased accordingly.

The change in magnetic entropy  $\Delta S(\bar{V})$

curve for a given  $\bar{V}$  and  $\sigma$  increases with increasing the volume exhibiting a maximum. With decreasing  $\sigma$  not only the position of the maximum  $\Delta S$  increases, but also the shape of the maximum becomes narrower. This behavior can be explained as by decreasing  $\sigma$ , the number of optimized volume is increased.

### f. The effect of dipolar interaction:

No observable effect seen, this is due to the negligible effect of the magnetic dipolar interaction term. Compared to other terms in the Hamiltonian.

### g. The effect of anisotropy constant:

With increasing the average anisotropy constant (K), the  $\Delta S$  decreases, this can be explained as follows, by increasing the energy barrier  $\Delta S$  is increased, or equivalently the effective volume is increased and hence the particles are easily aligned with the field, thus the  $\Delta S$  is increased.

## II. RELATED RESULTS

### (i) The effect of particle size distributions:

Fig.3.1 shows the entropy change for a non-interacting, spherically shaped superparamagnetic system. Assuming different distribution functions, the average diameter is (80 $\text{\AA}$ ), the standard deviation ( $\sigma$ ) for Gaussian (GD) and Log-normal (LD) distributions are  $7 \times 10^{-27}$  and 0.6 respectively, and the diameter range is (20-200 $\text{\AA}$ ). As the figure shows the  $\Delta S$  values for the uniform and Gaussian distributions are the same at any field.

The  $|\Delta S|$  values for (LD) a small increase over their corresponding values for (GD) or (UD) Sharp increases in  $|\Delta S|$  values are observed for rectangular distribution. Fig.3.2 Shows the entropy change for a non-interacting, spherically shaped superparamagnetic system. Assuming different distribution widths;  $10 < D_1 < 100 \text{ \AA}$ ,  $30 < D_2 < 80 \text{ \AA}$  and  $50 < D_3 < 60 \text{ \AA}$ . The  $|\Delta S|$  values are systematically increasing with increasing H, and the broader the width of the distribution is the larger the values of  $|\Delta S|$  are.

Fig.3.3 Shows the entropy change  $|\Delta S|$  for the same system used in Fig.3.1 and Fig.3.2 but assuming Gaussian distributions with different mean values of  $\bar{D}$  and standard deviations. The larger the average diameter is the larger  $\Delta S$  value at any H. Fig.3.4. Shows the  $|\Delta S|$  Vs T at  $H=5\text{kOe}$ , for the same system and conditions used in Fig 3.1. In each curve  $|\Delta S|$  decreases with increasing T, and the rate of decrease is slower as T increases.

The rate of decrease of  $\Delta S(T)$  with increasing T is almost the same for the Log-normal, Gaussian and Uniform distribution functions, being less rapid for Rectangular distribution function.

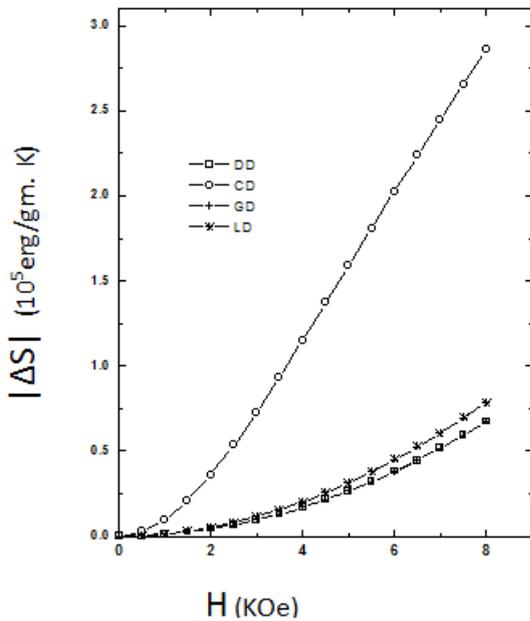


Fig 3.1 Shows  $|\Delta S|$  Vs  $H$  for all distribution without interaction with  $T=300$  K, the mean diameter is  $80 \text{ \AA}$ ,  $\sigma_3=7 \times 10^{-26}$ ,  $\sigma_4=0.6$ , the diameter range is  $(20-200 \text{ \AA})$ .

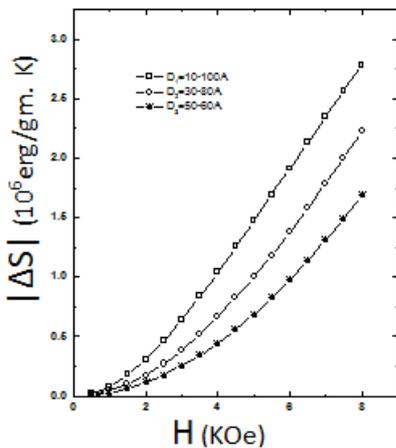


Fig 3.2 Shows the change in entropy  $|\Delta S|$  Vs  $H$  for CD at  $T=300$  K, the range of mean diameter is  $10 < D_1 < 100 \text{ \AA}$ ,  $30 < D_1 < 80 \text{ \AA}$ ,  $50 < D_1 < 60 \text{ \AA}$ .

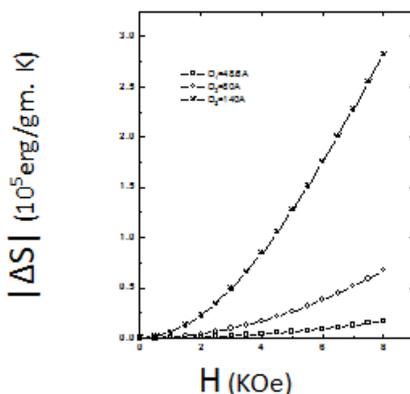


Fig 3.3 Shows  $|\Delta S|$  Vs  $H$  for GD without interaction with  $T=300$  K,  $D_1=48.6 \text{ \AA}$ ,  $D_2=80 \text{ \AA}$ ,  $D_3=140 \text{ \AA}$ ,  $\sigma_1=1.8 \times 10^{-26}$ ,  $\sigma_2=7 \times 10^{-26}$ ,  $\sigma_3=2 \times 10^{-25}$ , the diameter range is  $(20-200 \text{ \AA})$ .

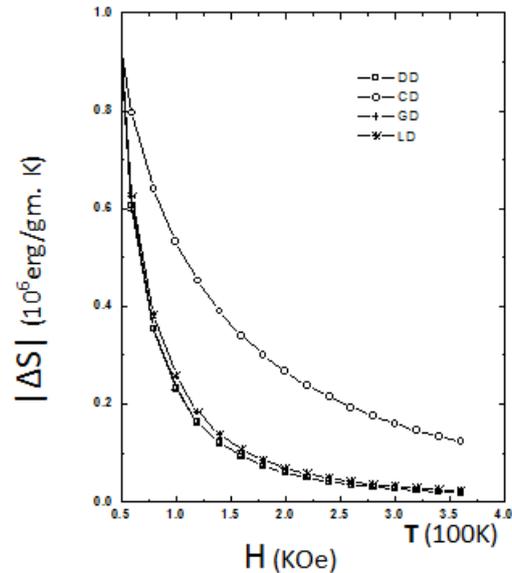


Fig 3.4 Shows  $|\Delta S|$  Vs  $T$  for all distribution without interaction with  $H=5$  kOe, the mean diameter is  $80 \text{ \AA}$ ,  $\sigma_3=7 \times 10^{-26}$ ,  $\sigma_4=0.6$ , the diameter range is  $(20-200 \text{ \AA})$ .

**(ii) The effect of standard deviation ( $\sigma$ ):**

Fig 3.5. Shows the  $|\Delta S|(T)$  curves for superparamagnetic system assuming the existence of Rectangular distribution function, with the same average diameter of particles ( $D=55 \text{ \AA}$ ), but with different widths.

The  $|\Delta S|(T)$  values decreases with increasing  $T$ , but at  $T=90$  K the three curves have the same  $\Delta S$  value, then the rate of decrease is reversed, from being rapid for the narrow distribution, for  $T \leq 90$  K, to being slower at  $T \geq 90$  K.

Fig 3.6. Shows the  $|\Delta S|(T)$  curves for superparamagnetic system assuming (GD) function with varying mean diameter values and standard deviation i.e.

$[(D_1, \sigma_1)=(48.6 \text{ \AA}, 1.8 \times 10^{-26})]$ ,  $[(D_2, \sigma_2)=(80 \text{ \AA}, 7 \times 10^{-26})]$  and  $[(D_3, \sigma_3)=(140 \text{ \AA}, 2 \times 10^{-25})]$  respectively. As the figure shows the highest mean diameter values shows the largest  $|\Delta S|$  changes.

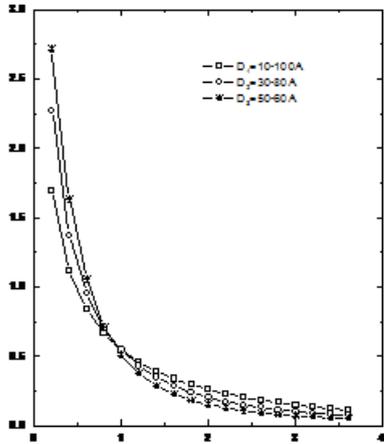


Fig 3.5 Shows the change in entropy  $|\Delta S|$  Vs the absolute temperature  $T$  for CD at  $H=5000$  Oe, the range of mean diameter is  $10 < D1 < 100$  Å,  $30 < D1 < 80$  Å,  $50 < D1 < 60$  Å.

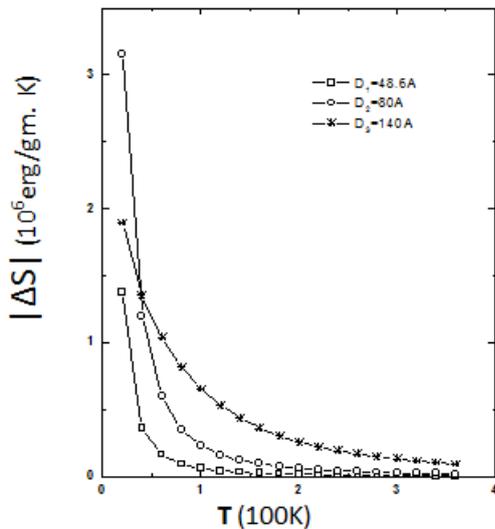


Fig 3.6 Shows  $|\Delta S|$  Vs  $T$  for GD without interaction with  $H=5kOe$ ,  $D1=48.6$  Å,  $D2=80$  Å,  $D3=140$  Å,  $\sigma1=1.8 \times 10^{-26}$ ,  $\sigma2=7 \times 10^{-26}$ ,  $\sigma3=2 \times 10^{-25}$ , the diameter range is (20-200Å).

**(iii) The effect of mean volume:**

Fig.3.7 Shows  $|\Delta S|$  curves for superparamagnetic systems as a function of mean volume for the Uniform, LN and GD

functions. For each distribution  $|\Delta S|(\bar{V})$  increases monotonically reaching a

maximum at  $\bar{V}=4.05 \times 10^{-24}$  GD(smooth one)

and at  $\bar{V}=2.05 \times 10^{-24}$  for LD, for the UD the

rate of increase of  $\Delta S$  with  $\bar{V}$  is fast at low

$\bar{V}$  then bends (and slower) for large  $\bar{V}$ .

Fig.3.8 Shows  $|\Delta S|$  versus  $H$  for a superparamagnetic system, assuming the existence of Log-normal distribution with different mean values and standard deviations,  $\Delta S$  increases with increasing  $H$  for each distribution but the higher the average value the higher the  $|\Delta S|$  values. Fig.3.9 Shows the  $|\Delta S|$  Vs average volume for a superparamagnetic system obeying Log-normal distribution at  $T=300$  k and  $H=5kOe$ , but varying the standard deviation  $\sigma$ . For each curve a maximum in  $|\Delta S|$  curve is observed, and the higher the

standard deviation the higher the value of  $\bar{V}$

at which maximum occurs, being at  $\bar{V}$

$=2.05 \times 10^{-24}$  for  $\sigma=0.6$ ,  $\bar{V}=3.3 \times 10^{-24}$  for

$\sigma=0.1$ ,  $\bar{V}=2.55 \times 10^{-24}$  for  $\sigma=0.3$ .

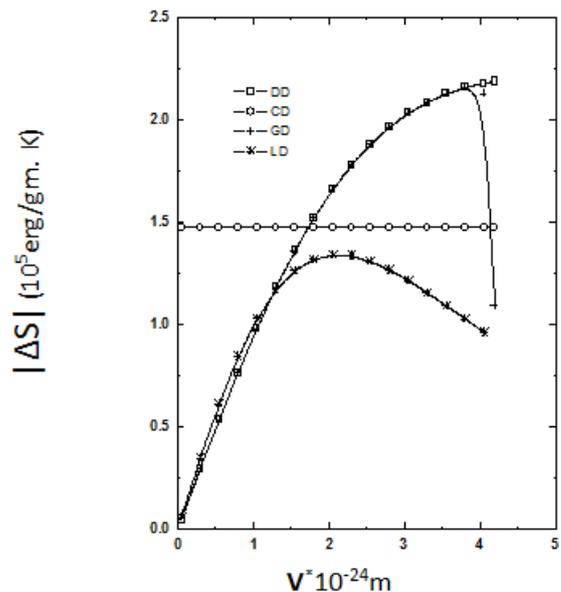


Fig 3.7 Shows  $|\Delta S|$  Vs  $V$  for all distribution without interaction with  $T=300K$  and  $H=5kOe$ ,  $\sigma3=7 \times 10^{-26}$ ,  $\sigma4=0.6$ , the diameter range is (20-200Å).

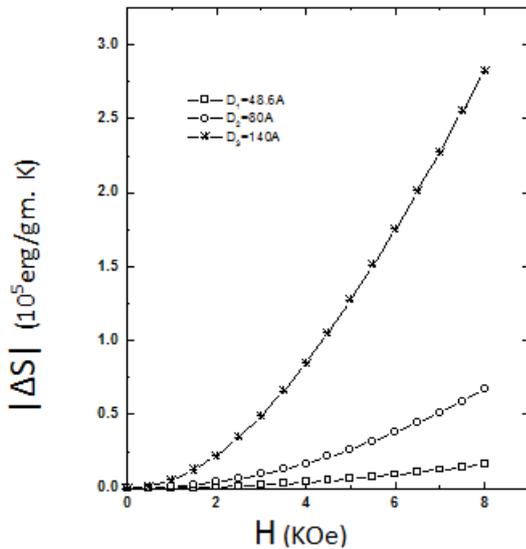


Fig 3.8 Shows  $|\Delta S|$  Vs  $H$  for LD without interaction with  $T=300K, D1=48.6 \text{ \AA}, D2=80 \text{ \AA}, D3=140 \text{ \AA}, \sigma1=0.6, \sigma2=0.1, \sigma3=0.3$ , the diameter range is (20-200Å).

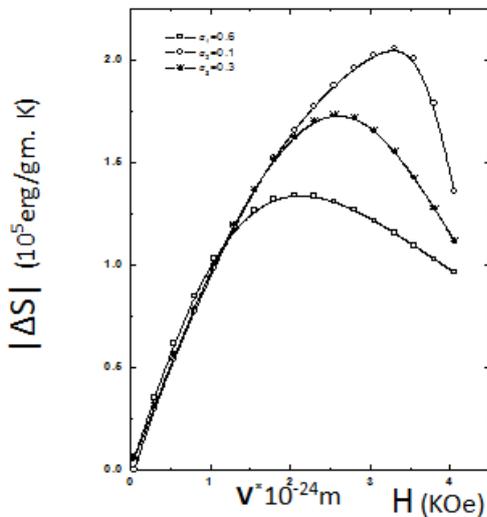


Fig 3.9 Shows  $|\Delta S|$  Vs  $V$  for LD without interaction with  $T=300K$  and  $H=5kOe, \sigma1=0.6, \sigma2=0.1, \sigma3=0.3$  the diameter range is (20-200Å).

**(iv) The effect of magnetic interactions:**

Fig.3.10 Shows the  $|\Delta S|(T)$  curves for the same parameters used in obtaining Fig.3.4, except, we add the contribution of magnetic interaction (the calculations were carried out using Eq(2-46)). The effect of the magnetic interaction is so small, that almost the same result as in Fig.3.4 was obtained.

Fig.3.11 Shows the  $|\Delta S|(T)$  curves for the same parameters used in obtaining Fig.3.6, except, we add the contribution of magnetic interaction (the calculations were carried out using Eq(2-46)), and assuming LD. The effect of the magnetic interaction is so small; again the same result as in Fig.3.6 was obtained.

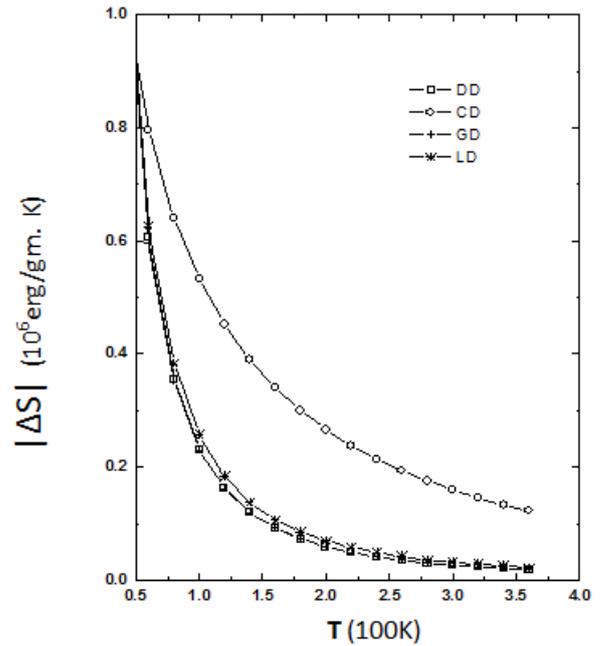


Fig 3.10 Shows  $|\Delta S|$  Vs  $T$  for all distribution with magnetic dipolar interaction with  $H =5kOe$ , the mean diameter is  $80 \text{ \AA}, \sigma3=7 \times 10^{-26}, \sigma4=0.6$ , the diameter range is (20-200Å).

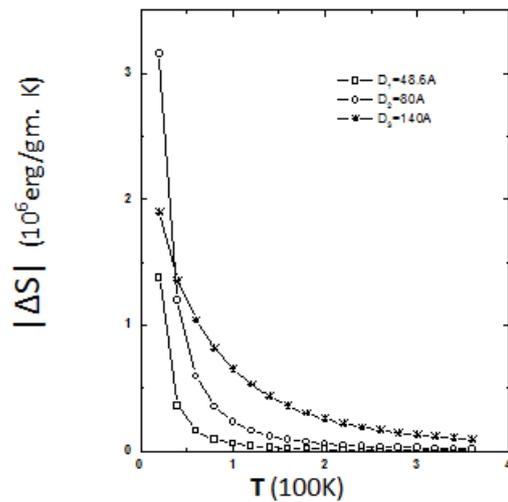


Fig 3.11 Shows  $|\Delta S|$  Vs  $T$  for LD with magnetic dipolar interaction with  $H=5kOe, \sigma1=0.6, \sigma2=0.1, \sigma3=0.3$  the diameter range is (20-200Å).

**(v) The effect of average anisotropy constant:**

Fig.3.12 Shows  $|\Delta S|$  versus H for a superparamagnetic system, assuming the existence of G-distribution with different mean values and standard deviations, we add the contribution of anisotropy constant (the calculations were carried out using Eq(2-34)), assuming (GD) function with mean diameter values and standard deviation i.e.

$[(D, \sigma)=(50\text{\AA}, 0.6 \times 10^{-26})]$  and the anisotropy constant are  $K_1=5\text{kJ/gm.K}$ ,  $K_2=10\text{kJ/gm.K}$ , and  $K_3=15\text{kJ/gm.K}$ .  $|\Delta S|$  increases with increasing H for each distributions but the higher the anisotropy the higher the  $|\Delta S|$  values.

Fig 3.13. Shows the  $|\Delta S|(T)$  curves for superparamagnetic system assuming the existence of G-distribution with different anisotropy constant, we add the contribution of anisotropy constant (the calculations were carried out using Eq(2-34)). assuming (GD) function with mean diameter values and standard deviation i.e.

$[(D, \sigma)=(50\text{\AA}, 0.6 \times 10^{-26})]$  and the anisotropy constant are  $K_1=5\text{kJ/gm.K}$ ,  $K_2=10\text{kJ/gm.K}$ ,  $K_3=15\text{kJ/gm.K}$ . As the figure shows the highest anisotropy constant values shows the largest  $|\Delta S|$  changes.

Fig 3.14. Shows the  $|\Delta S|(K)$  curves for superparamagnetic system assuming (GD) function with varying mean diameter values and standard deviation i.e.  $[(D_1, \sigma_1)=(30\text{\AA}, 1.414 \times 10^{-26})]$ ,  $[(D_2, \sigma_2)=(50\text{\AA}, 6 \times 10^{-26})]$  and  $[(D_3, \sigma_3)=(70\text{\AA}, 2.5 \times 10^{-26})]$  respectively. we add contribution of anisotropy constant (the calculations were carried out using Eq(2-34)).

In each curve  $|\Delta S|$  increases with increasing K. and the rate of increase is slower as K increases for  $[(D_1, \sigma_1)=(30\text{\AA}, 1.414 \times 10^{-26})]$ ,  $[(D_2, \sigma_2)=(50\text{\AA}, 6 \times 10^{-26})]$ , but for  $[(D_3, \sigma_3)=(70\text{\AA}, 2.5 \times 10^{-26})]$  a rapid increase is observed. As the figure shows the highest mean diameter values shows the largest  $|\Delta S|$  changes.

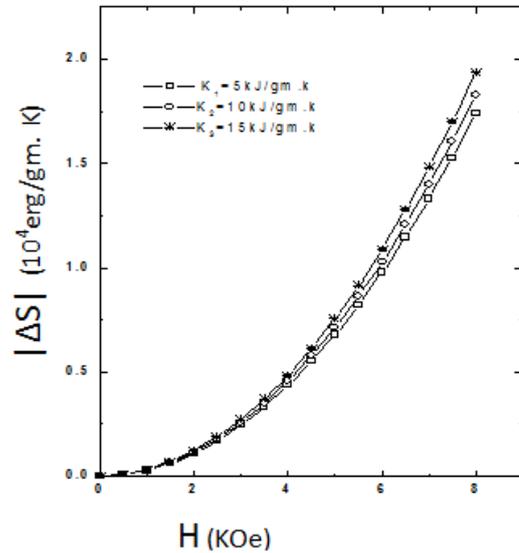


Fig.3.12 Shows  $|\Delta S|$  versus H for GD with anisotropy constant with mean diameter values and standard deviation i.e.  $[(D, \sigma)=(50\text{\AA}, 0.6 \times 10^{-26})]$ , the diameter range is  $(20-80\text{\AA})$ .

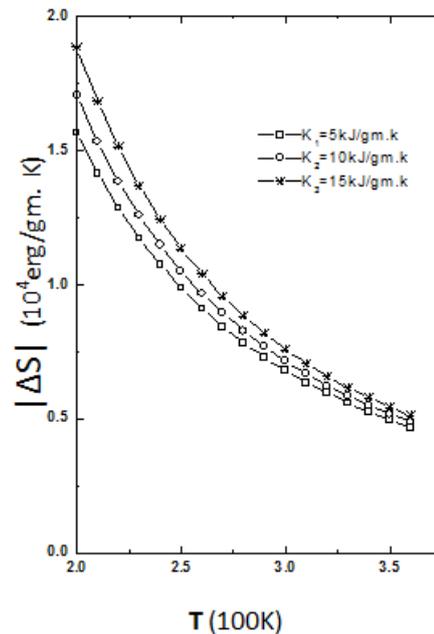


Fig.3.13 Shows  $|\Delta S|$  versus T for GD with anisotropy constant with mean diameter values and standard deviation i.e.  $[(D, \sigma)=(50\text{\AA}, 0.6 \times 10^{-26})]$ , the diameter range is  $(20-80\text{\AA})$  ..

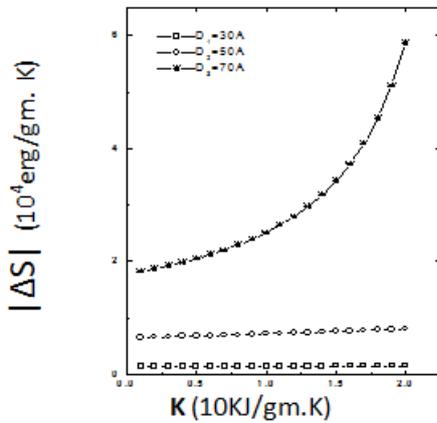


Fig 3.14. Shows  $|\Delta S|$  Vs  $K$  assuming (GD) function with anisotropy constant with varying mean diameter values and standard deviation i.e.  $[(D1, \sigma1)=(30\text{\AA}, 1.414 \times 10^{-26})]$ ,  $[(D2, \sigma2)=(50\text{\AA}, 6 \times 10^{-26})]$  and  $[(D3, \sigma3)=(70\text{\AA}, 2.5 \times 10^{-26})]$  respectively. the diameter range is (20-80 $\text{\AA}$ ).

#### IV CONCLUSION:

In the presented paper provides the thesis work based on the effect of particles

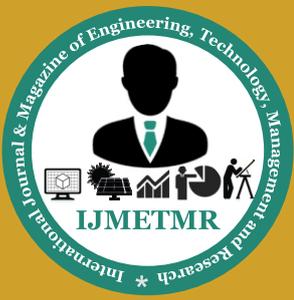
In the presented paper provides the thesis work based on the effect of particles size distributions  $F(V)$  on the magnetic entropy change  $|\Delta S|$  in magnetic fine particles system which were calculated numerically as a function of mean volume of

particles ( $\bar{V}$ ), the standard deviation ( $\sigma$ ), the magnetic field ( $H$ ), and the absolute temperature ( $T$ ). For each  $\Delta S$  curve the results were compared with the corresponding results obtained using the classical theory of superparamagnetism. Four distribution functions were taken Uniform distribution (UD or DD), Constant distribution (CD), Gaussian distribution (GD) and Log-Normal distribution (LD). It is clear that with the exception of rectangular distribution (which is not suitable for ultra fine particle systems), the magnetic entropy change is not affected by the kind of distribution. This due to the fact

that there is a certain particle size, which contribute significantly to the magnetic entropy change. For the case of rectangular distribution, large portion of particles lie in the size range which corresponds to the maximum entropy change, therefore, using the RD corresponds to  $\Delta S$  greater than their corresponding using GD or LD.

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