The 6th International Conference on Manufacturing Research (ICMR08)
Brunel University, UK, 9-11th September 2008

HIGH ENERGY MILLING OF MICRO MAGNETIC POWDER FOR FINGERPRINT DEVELOPMENT

K. Nag [1], X. Liu [1], A. Scott[2], Y. K. Chen [3]

1. School of Computing, Engineering and Physical Sciences, University of Central Lancashire, UK
2. School of Forensic and Investigative Sciences, University of Central Lancashire, UK
3. School of Aerospace, Automotive and Design Engineering, University of Hertfordshire, UK

Abstract

Highly reflective magnetic powders have been available commercially for latent fingerprint development on dark background surfaces for a few years, however there are needs of a superior darker variety of the magnetic powder which would be ideally suitable for obtaining good contrast on light background surfaces. A novel dark magnetic powder is therefore suggested for the application in latent fingerprint development on light backgrounds. Based on a comprehensive analysis of the manufacturing techniques for the production of metallic powders and previous experiences, a series of dry milling trials were proposed using a high energy vibratory mill. In these milling trials, atomized iron powders of appropriate particle dimensions were chosen as the starting material. The starting atomized iron powders are mixed with a specific process controlling agent in cylindrical plastic pots, which are fixed on the vibratory mill. Stainless steel balls were used as the milling medium. The amount of starting iron powders and the milling time are designated so as to obtain flakes of different particle dimensions. After the designated milling process, the powders were carefully exposed to the air to avoid catching fire and separated from the steel balls. Thereafter, the samples of the powders were inspected using scan electron microscope and the result reveals that some good quality flakes with leafy characteristics and high aspect ratio were developed. The samples of the powders were also used to develop latent fingerprints on a common background at the university's forensic investigation laboratory, and the result indicates that some flakes obtained can be considered as good quality darker variety of magnetic powder for fingerprint development.

Keywords: High Energy Milling, Magnetic Iron Flakes, Fingerprint Powder.

HIGH ENERGY MILLING OF MICRO MAGNETIC POWDER FOR FINGERPRINT DEVELOPMENT

1.0 Introduction

Metallic flake powders have been widely used for fingerprint detection. In the UK there are several varieties of fingerprint powders available from different forensic suppliers, although very few of these powders are actually manufactured solely for the purpose of fingerprint detection. Typical examples of flake powders which are used extensively for fingerprint detection are aluminum powder, brass powder and Magneta Flake (a commercial variety of magnetic flake). Highly reflective aluminum powder has been the most widely used powder for fingerprint detection in crime scenes investigation and is generally employed for bright fingerprint development on dark backgrounds (most effective on glass surface). However with the introduction of Magneta Flake in the last decade, high density magnetic flake (e.g. iron) when applied on crime scenes, has been able to overcome the problem of air-borne dust level typically associated with application of low density aluminum flake when applied with standard squirrel brush [1]. Also because of its low density, aluminum powder tends to take some time to settle during application. Furthermore, the magnetic flakes are applied on the crime scene with a magnetic applicator thereby leading to somewhat non destructive fingerprint detection. Magneta Flake thus, produced a suitable alternative and as with aluminium flake it is found to be extremely reflective and ideally suitable for dark backgrounds. Recent home office evaluation of various fingerprint powders suggest that Magneta Flake actually outperforms aluminum on lot of surfaces like painted metal, gloss painted wood and others and closely matches the performance of aluminum on glass surfaces [2]. While Magneta Flake has been successfully used for bright fingerprint development on darker backgrounds, there is not a suitable powder obtained commercially which could be as effective as Magneta Flake on lighter backgrounds. Further, Home Office report suggests that one of the black magnetic powder varieties obtained commercially is actually a mixture of two particles, one large magnetic carrier particles of iron (20-200 μm) and the other smaller non magnetic particles of iron oxide (3-12 μm) which actually develops the mark in adhering to the fingerprint residue. It is however, the least used powder in crime scenes investigations, and is not very effective on most surfaces as compared to that of aluminum powder and Magneta Flake. Previous research with different metal flakes suggests that iron flake with diameters 10-25 μm would allow print development to a quality considerably superior to that of other commercial black and magnetic black powders, when applied by a magnetic applicator on light backgrounds [3]. However, manufacturing of such powders had found little success over the years. The aim of the present study is therefore to develop a superior quality of dark iron magnetic flake fingerprint powder suited for lighter backgrounds on a wide range of surface textures. A set of initial experiments have therefore been conducted to develop the darker variety of magnetic flakes and subsequently been analyzed for its suitability as a fingerprint powder.

2.0 Factors Governing the Development of Dark Flake for Forensic Applications

Although most of the commercially available metal flakes including aluminum flakes are usually produced by rotary ball milling in tonnage quantities**,** various high energy milling devices are often utilised for rapid production of trial quantities of metal powders for experimentation. They differ mostly in terms of their design and modes of operation. However, the fundamentals of change in particle morphology of ductile metal powder during high energy milling is same for all the devices and can be attributed to a combination of phases, i.e. *micro-forging*, *fracture*, *agglomeration* and *de-agglomeration*, all of them can take place simultaneously in a mill [4]. It is therefore important that periodic samples are drawn at different stages of a milling experiment and analysed in order to identify the different milling phases.

2.1 Role of Additives as a Process Controlling Agent (PCAs)

Although a large number of solid or liquid additives are used in practice in different milling applications, stearic acid ($CH_3(CH_2)_{16}COOH$) is the most common PCA used in metal flake pigments for both laboratory purposes and industrial applications. The most important function of stearic acid as an additive is essentially to lubricate the powder particles, thereby helping the process of microforging of flakes. In absence of stearic acid, frictional forces prevent particles sliding over each other and a considerably large number of particles will be involved in the impact between the balls. With addition of stearic acid, it allows particles to flow over one another resulting in fewer particles coming under the entrapment zone. Thus fewer particles will receive the impact energy and will have a greater strain increment, thereby initiating microforging. Further, studies have indicated that milling of aluminum under oxygen without stearic acid has resulted in the powder remaining in the granular form and showed a slight increase in apparent density. With the usage of stearic acid there was a substantial fall in both in apparent density and oxygen pressure, indicating that flakes have been formed with high surface area of aluminum reacting with oxygen. This has been explained by the mechanism of interaction between stearic acid and the metal in formation of the flakes. Essentially a metal stearate is formed in which metal atom is bound in the surface and the stearate atoms are roughly oriented perpendicular to the surface. Further, layers of free stearic acid may be formed above this layer which provides resistance to cold welding of the metal particles thereby inhibiting agglomeration. Another important role of stearic acid in production of fingerprint powder is that the thin coat of stearic acid remaining on the flake surfaces helps in the adhesion of flakes to a latent fingerprint deposit. Previous studies have shown that removal of stearic acid coating by solution in a suitable solvent (usually soxhlet in hot acetone) seriously reduces the effectiveness of flakes for fingerprint development [5]. It is to be mentioned that in previous studies with fingerprint powders, a variety of alternate organic coatings have been experimented with, particularly with substances which are secreted in sweat and/or sebum present in latent fingerprint residues (e.g. tripalmitin, tristearin, squalene) [6] but none of them seemed to match the quality achieved by stearic acid.

2.2 Choice between Wet and Dry Milling

Two types of milling environment can be defined based on the formation of surface films on the metal powders, reactive and non reactive milling. In reaction milling the powder surface reacts extensively in the fluid to produce surface films which inhibits agglomeration by welding. This would result in the milled powder being extremely fine. In non reactive milling, the powder particles hardly react with the milling fluid, thereby bare metal surfaces are formed which enhances the weldment of the powder particles. Metal powders milled in organic or inorganic fluids retain small amounts of the fluid dispersed throughout each particle. Thus, hydrocarbons containing hydrogen and carbon and carbohydrates containing hydrogen, carbon, and oxygen are likely to introduce carbon and/or oxygen into the particle. In essence, in wet milling a milling liquid can interact with the metal particle and influence in the same way a gaseous medium would interact in dry grinding. Normally when the final product is dry, dry milling is preferred as in the case of producing metal flakes for most industrial applications. Wet milling of metal particles is considered only when flakes fail to form by conventional dry milling route. It is therefore suggested that dark fingerprint powder be produced by dry milling, wet milling with suitable liquid can only be considered if the dry milling route cannot yield any positive results.

HIGH ENERGY MILLING OF MICRO MAGNETIC POWDER FOR FINGERPRINT DEVELOPMENT

2.3 Colour and Visual Quality of the Powder

One of the important criteria of developing a fingerprint powder is to understand how we can obtain the desired colour of the powder. The colour of the metal flake can be governed by two factors, the basic colour of the starting material used and the presence of surface films on the metal surface. The basis colour of the metal surfaces arises out of the interaction of the electric field of light waves with conduction electrons of the metal; the detailed phenomenon is not discussed here. Essentially it can be said that to obtain a dark fingerprint powder, the colour of the starting material should not be bright and preferably darker in appearance. However, during the milling operation the starting material may undergo variety of shape changes at different stages of milling and would therefore exhibit different colours due to either specular or diffuse reflection of light. It is expected that a relatively equiaxial particle will reflect light in a diffuse manner where as flat flakes will tend to be glossy due to the specular reflection. Further the flakes should be deposited parallel to each other (leafing)[7], if not, the flakes will scatter the light by overall diffuse reflection and specular reflection will take place over very small distances of the order of the flake diameter, which would result in the loose flakes appear to sparkle. Further, although flat parallel flake surfaces would give specular reflection, the edges/perimeter of the flake will still be radiating at different angles (diffuse reflection). Thus with decreasing flake diameter the edge effect will tend to predominate and there will be more diffused reflection. It is therefore expected that to obtain dark fingerprint powder, the flake diameter has to be small, although optimum size has to be determined based on the quality of developed fingerprints. Another important factor which governs the colour of the final product is the surface chemistry of the flakes i.e. either a presence of oxide layer, a layer of metal stearate or free stearic acid; all of which are encountered in the milling process. Thickness of the oxide layer can influence the colour of the product particularly when the milling environment is non inert, i.e. in presence of air. The level of the metal stearate and/or free stearic acid could also influence the flake colour as interference effect could be produced if the layers are thick enough.

3.0 Experimental Procedure

The choice of an appropriate starting material has been determined based on previous studies [8], where it has been observed that very fine powder like iron carbonyl when milled by standard dry grinding method, forms a solid mass by agglomeration and so wet milling in a suitable liquid had to be introduced for the formation of flakes. Since the study also suggested that small particle size of the starting powder is beneficial to the production of flakes by high energy milling, in the present study, atomized iron powder manufactured by Sigma Alldrich, of approx average particle size of 15 μm has been selected as the starting material. Also the powder selected is irregular in shape as flakes produced from irregular shaped powder tend to produce dark flakes as compared to spherical particles which produce highly reflective flakes like that of Magneta Flake. A set of experiments have been carried out with different amount of starting material in a prototype vibratory mill at 3000 rpm, 50 Hz. The starting material along with 2-4 wt% of stearic acid as process controlling agent are charged in plastic containers and milled intermittently for a length of time with the aid of 7mm stainless steel balls. The milling time, the amount of starting material varied during the dry milling process to obtain flakes of different particle dimensions. Iron flake powder of 1g during the milling was sampled and characterized, subsequently analyzed by scanning electron microscope. Remaining powder was utilised for developing latent fingerprint development. Since the quality of the print can vary depending on the level of fingerprint residue, a standard procedure was therefore adopted to obtain a set of virtually identical

fingerprints. A single donor rubbed his hand to distribute sweat over his fingers before pressing all his fingers on a white piece of paper. The same process was repeated for deposition of all the prints. The fingerprints were then developed for each of the powder samples using a magnetic applicator and subsequently they were scanned directly in the university's forensic department with the aid of 'Livescan', a device used for AFIS (Automated Fingerprint Identification System) in crime scene investigation.

4.0 Results and Discussion

4.1 Milling Behavior of the Iron Powder and Feasibility of the Dry Milling Process

The milling of the powder has been carried out in a prototype vibratory mill which can generate great impact forces due to the rapid vibration of the motor thus resulting in faster and finer grinding. The starting atomized powder has been milled for a maximum of 9 hrs and it has been observed that flakes of 25-30 μm have been obtained after milling for at least 4 hrs, depending on the amount of starting material used. Fig 1 shows the scanning electron microscopic image of the starting material which during the course of milling has undergone a combination of milling phases, primarily microforging and fracture. The irregularity of the particle shape of the resultant flakes indicate that the starting irregularly shaped particles are actually compressed in a non uniform manner which has resulted in flakes with complex surface contours and jagged outlines.

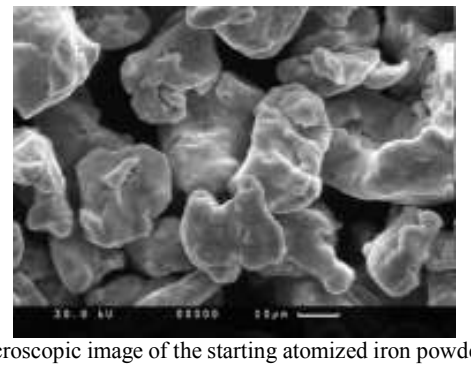


Fig 1. Scanning Electron Microscopic image of the starting atomized iron powder manufactured by Sigma Alldrich

During the experimental study it has been observed that the dry milling process in producing flakes is feasible only when the process is carried out with some careful considerations. The powder flakes exhibited pyrophoric behavior and tend to ignite spontaneously when being exposed to atmosphere. The milling containers, made of thin plastic did not heat up substantially during the milling process and the powders only starts burning once the milling vials are opened and the flakes are being exposed to atmosphere. The burning is in the form of spatters, starting from few powder particles, then progressively spreads to other powder particles to form a lump at the end, unless they are interrupted in between and separated. However, it has been observed that with controlled exposure of powder to the atmosphere by slow bleeding of air into the milling vials at the end of the milling process and carefully separating the powder from the balls, the powder tends not to burn in atmospheric conditions. It is therefore suggested that to negate the effect of oxidation of the powder particles, dry milling should be carried out in an inert atmosphere with the addition of 2-5% of oxygen to allow careful oxidation within narrow limits. This would enable the formation of protective oxide films on the flakes thereby reducing the pyrophoric behaviour of the powder particles.

4.2 Effect Of Milling Time And The Amount Of Starting Material Used

Two sets of experiments were carried out with different amount of starting material; the first set of experiment with 5g of powder and the second experiment with amount varying between 15-30g. Milling time has been varied between 4-9 hrs in order to achieve flakes of different dimensions. Typically, flakes of average particle dimensions of 25-30 μm and 10 μm have been obtained as previous studies have suggested that such flake dimensions can be considered as optimum for latent fingerprint development.

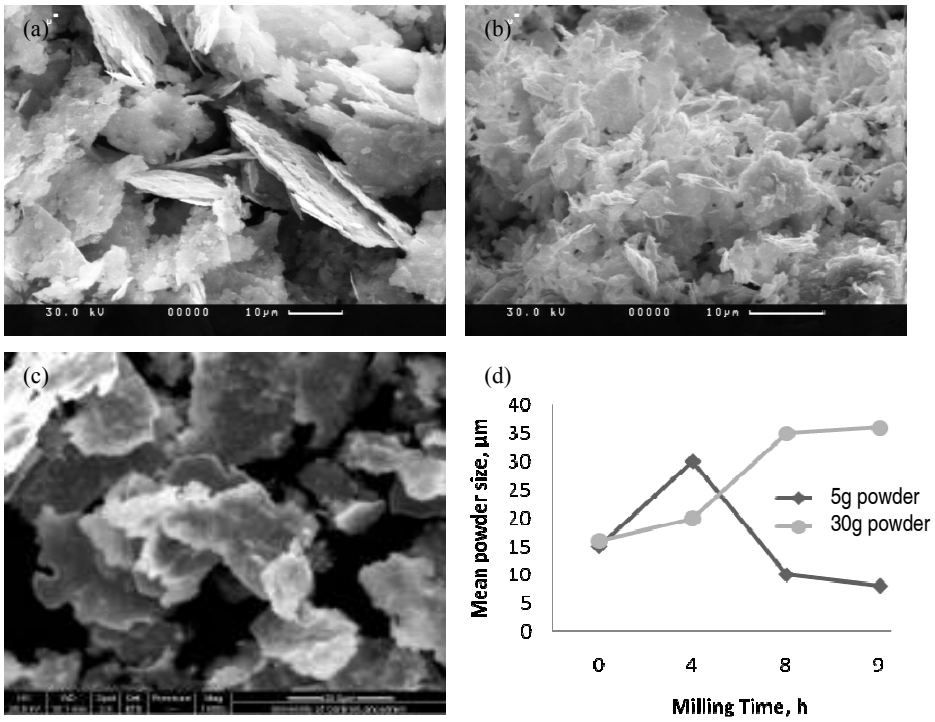


Fig 2. Scanning Electron Microscopic image of dark iron flakes when starting material of quantity (a) 5g is milled for 4hrs (b) 5g is milled for 8 hrs and (c) 30g is milled for 8hrs and (d) graph showing the effect of milling time and the amount of starting material used (5g and 30g with similar ball to powder weight ratio) on the mean flake dimensions.

It has been observed that with 5g of starting material, dry milling for 4 hrs have resulted in flakes of average particle size of 25-30 μm. Figure 2a shows a scan electron microscopic image of a typical sample of dark powder from the 1st set of experiments, after being milled for 4 hours. The particles as observed in Scan Electron Microscope (SEM), show good flaky characteristic, but is not uniform throughout. It showed the presence of bigger particles which have not been milled properly. However, more consistent flake sizes have been obtained when a sample of the resultant flakes has undergone sieving operation and +38 μm of the fraction is being discarded. A different sample, under identical set of milling condition having undergone milling for 4 hours is being milled further for another 15mins with the aid of glass beads to achieve more uniform flakes. However, it did not show any marked difference in the surface characteristics of the flake. Further milling of powder flakes up to 8hrs has resulted in achieving flake dimensions <10 μm. Figure 2b shows a scan electron microscopic image of a typical sample of dark powder from the 1st set of experiments, having undergone milling for 8 hours. SEM analysis of the powder reveals consistent flakes have been obtained with very good leafy structures and with relatively uniform distribution.

HIGH ENERGY MILLING OF MICRO MAGNETIC POWDER FOR FINGERPRINT DEVELOPMENT

In the initial stage of the milling process, the particles are compressed via repeated impact of the milling medium which results in a deformation (flattening) of the particle. Thus good quality flakes have been obtained of required particle dimension after milling for 4hrs. Further milling of the flakes has resulted in increased fracture of the particles due to the initiation and propagation of cracks across the particle. The cracks may occur due to work hardening, ductile rupture as the flake becomes thinner in some parts than in others, fatigue failure, fragmentation, initial defects and inclusions in particle, or a combination of these effects. This has resulted in decreasing the particle dimension to <10 μm after 8 hrs of dry milling. During the milling process, it has been observed that the amount of starting material has pronounced effect on the milling pattern. While 25-30 μm particle size has been obtained by milling 5g of powder for 4hrs, almost 8 hrs of milling was required for 30g of powder to achieve the same results. Figure 2c shows a scan electron microscopic image of a sample of dark powder from the 2nd set of experiments, where 30 g of powder has undergone 8 hrs of milling. The sample after being milled for 4 hrs did not sufficiently flatten the powders to form flakes although there has been a noticeable increase in diameter to thickness ratio. With further milling of the powder, it shows the tendency of microforging and subsequent initiation of cracks until an optimum size 25-30 μm flakes have been obtained after 8 hrs of milling. Fig 2d shows the variation in mean powder size of two samples with respect to the milling time for 5g and 30g of starting material used. Other samples with 15-25 g of powder also took much longer than 4 hrs of milling to obtain the required flake dimensions. These results clearly demonstrate that as the quantity of the starting powder charged into the milling vials are decreased, the processes of microforging, fracture and agglomeration occurred more rapidly. In all the cases, milling process was not carried out beyond 9 hours as it has been decided that total production time should not exceed 10 hours (including set up time, time for charging and discharging etc.) to make the process viable for rapid production of metal flakes on a laboratory scale. As a result, it was not possible to obtain flakes of 10 μm variety with 20-30 g of powder. It is however, expected that longer milling time up to 16 hrs would be required to obtain 10 μm variety of flakes under the present experimental set up. The overall milling time can however be reduced by changing and optimising the process parameters like increasing the efficiency of the vibratory mill, optimising the ball to powder ratio, size of the balls and quantity of the stearic acid used. The slight variation of the stearic acid level between 2-5 wt% under present experimental conditions however, had negligible effect on the milling time as well as the quality of flakes produced.

4.3 Comparative Analysis with Magneta Flake & Suitability for Latent Fingerprint Development

Samples from two set of experiments have been visually inspected and digitally photographed to compare the coloration and appearance of the powder which would be an important factor when powders are to be applied on common backgrounds to test the suitability for latent fingerprint development. It has been observed that flakes of particle dimension less than 10 μm is much darker in appearance when compared to that of 25-30 μm flakes. This can be attributed to the fact that due to the smaller average particle size of sample, edge effect is more predominant which contributed to more diffused reflection of light, as explained earlier. A sample of commercially available Magneta Flake has also been analysed in SEM and a comparative analysis has been made with the dark powder produced, as briefed in Fig 3. Elementary analysis of the powders as well as the starting material, reveals that the dark powder has got higher oxygen content compared to that of Magneta Flake, where it is almost negligible. This indicates that the dark powder has undergone slight oxidation during the course of dry milling as the starting material has found to be of pure iron without presence of any oxides. It is further corroborated from the fact that during the experiments, it was difficult to open the covers of the plastic vials indicating that there is a pressure drop inside as a result of the slight oxidation.

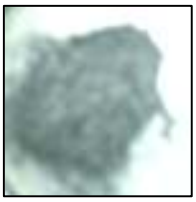
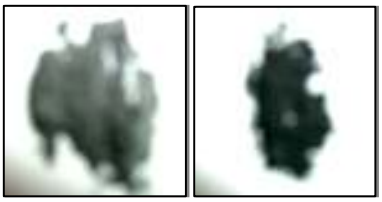
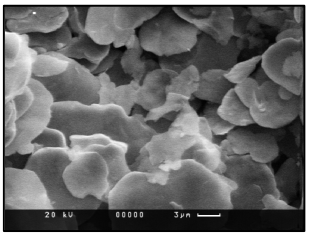
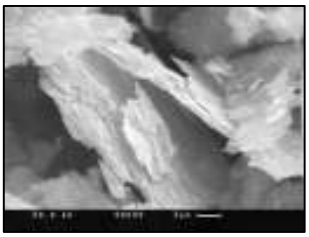
Particle	Magneta Flake	Dark Powder
Composition	Iron flake	Iron flake
Process	Wet Milling	Dry Milling
Description	Flat structure, smooth surface Highly reflective	Rough surface with jagged edges Dark appearance
Distribution	Uniform	More or less uniform
Grouping	Moderate	Moderate
Thickness	Thin	Extremely thin
Particle Diameter	10-15µm	25-35 µm, <10µm
Visual Characteristics for a lump of powder		
Typical Scanning Electron Microscopic Image		

Fig 3. Comparative analysis between dark flake powder and bright magnetic flakes (Magneta Flake)

A set of samples from both experiments have been utilised for developing latent fingerprints on a white background following a standard procedure to obtain identical fingerprints. Since the magnetic flakes are specifically useful for latent print development on porous surfaces such as wall, paper and polythene [9]; 80gsm A4 sized white papers have been considered for impression and development of latent fingerprints. A commercial variety of black magnetic powder has also been included in the analysis to identify the quality of fingerprint developed with respect to that achieved by the dark flakes. Analysis of the developed prints reveal that good quality of prints has been obtained using both the 25-30 µm and 10 µm variety of the flakes. Fig 4. shows the scanned images of the latent fingerprint developed by the three types of powders considered. It is evident that the best quality of developed prints has been obtained by 25-30 µm variety of the flakes with good ridge details and reasonably good contrast. It has been observed that 10 µm variety of the flakes also allowed reasonably good quality of print development with much darker appearance producing a better contrast against a white background. However, this fine flake tends to paint over the surface of the paper, thereby somewhat reducing the quality of the overall print developed. In either case, the print developed on white paper background has found to be much superior to that obtained by the commercially available magnetic powder. Further analysis by the Automated Fingerprint Identification System (AFIS) in the university's forensic department also conform the findings. It is to be mentioned that the print quality did vary slightly from sample to sample indicating that best fingerprint quality can be achieved by optimising the flake characteristics and particle dimensions. Further, it is also suggested that similar experiments can be conducted with other types of iron/steel powders as starting materials and find their potential as magnetic flakes for fingerprint development.

HIGH ENERGY MILLING OF MICRO MAGNETIC POWDER FOR FINGERPRINT DEVELOPMENT

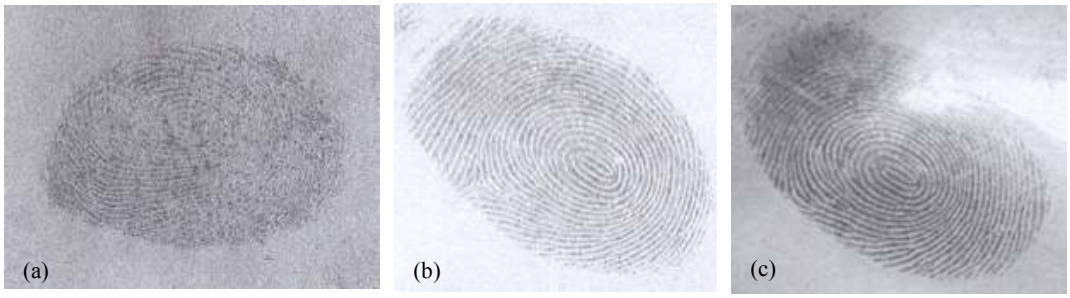


Fig 4. Quality of fingerprints developed under identical set of conditions on a white paper using a magnetic applicator by (a) commercially available magnetic fingerprint powder (b) 25-30 µm dark iron flakes and (c) 10 µm dark iron flakes

5.0 Conclusions

High Energy Milling devices, particularly a vibratory mill can be used for rapid production of micro magnetic metal flakes which has potential applications in fingerprint technology. The magnetic properties of iron flake together with the flake characteristics achieved by dry milling can be utilised for developing a suitable darker variety of magnetic flakes for latent fingerprint development on light backgrounds. Analysis of these flakes produced from a set of initial experiments reveal that print developed can easily surpass the quality achieved by commercially available dark magnetic powders on porous surfaces. The quality of the flakes can be further enhanced by optimising the dry milling parameters including the amount of starting material, milling time and other criteria like size of milling medium, amount of process controlling agents etc. Further, other varieties of dark magnetic flakes can be produced from different starting material which can be suitable for developing latent fingerprints on a wide range of background surfaces.

References

[1] J. D. James, C. A. Pounds, and B. Wilshire, "Magnetic flake fingerprint technology," *Journal of Forensic Identification*, Vol. 41, No. 4, 1991, pp 237-247.
[2] Helen L. Bandey, Andrew P Gibson, "The powders process, study 2: evaluation of fingerprint powders on smooth surfaces," *Fingerprint Development and Imaging Newsletter*, Pub. No. 08/06, Feb 06.
[3] James, J. D., Pounds, C. A., and Wilshire, B., "Flake Metal Powders for Revealing Latent Fingerprints," *Journal of Forensic Sciences*, JFSCA, Vol. 36, No. 5, Sept 1991, pp 1368-1375.
[4] W. E. Kuhn, I. L. Friedman, W. Summers, A. Szegvari, "Milling of brittle and ductile materials", *ASM Handbook*, *Powder metallurgy*, Vol 7, 1985.
[5] J. D. James, and B. Wilshire, "New powder metallurgy techniques for fingerprint detection," *Key Engineering Materials* (Switzerland), Vol. 86-87, 1993, pp 11-16.
[6] J. D. James, R. J. Gowland, and B. Wilshire, "Research yields new uses for commercial powders," *Advanced Materials Technology/International*, 1992, pp 94-99.
[7] R. Besold, H. C. Neubing, and E. D. Lloyd, "Metal flakes: highly innovative powder products," *Powder Metallurgy*, Vol. 32, No. 1, 1989, pp 25-28.
[8] J. D. James, C. A. Pounds, and B. Wilshire, "Production and characterisation of flake metal powders for fingerprint detection," *Powder Metallurgy*, Vol. 34, No. 1, 1991, pp 39-43.
[9] G.S. Sodhi, J. Kaur, "Powder method for detecting latent fingerprints: a review," *Journal of Forensic Science International*, Vol. 120, Issue 3, 2001, pp 172-176