High Performance Field Emitters

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ABSTRACT

By compiling the largest meta-analysis to date, across a diverse range of materials, here we show, for the first time, that the work function of a material does not strongly impact on field electron emission performance, when only the work function is considered. Materials studied include 1D, 2D and 3D/bulk with a range in work function from 1.33 eV to 7.75 eV. We find that the turn-on electric fields show a stronger dependency on dimensionality than on work function, with 3D/bulk materials typically requiring an electric field twice as large as those of 1D and 2D emitters.

INTRODUCTION

Cold cathode field emission from nanomaterials is an on-going area of great academic and technological interest. There have been many suggested applications of field electron emission, including displays, [1-3] [3] travelling wave tubes, [4, 5] [5] microwave amplifiers, [6, 7] [7], electron microscopy,[8, 9] [9] parallel electron beam lithography, [2, 10, 11] and X-ray sources. [2, 12, 13] [13] [10]. Low work functions have been repeatedly touted as primary drivers towards achieving high performance field electron emitters [14, 15] [15]. However, detailed analysis of the way in which this characteristic affects the field emission has not yet been fully understood or comprehensively studied across a diverse range of materials. Supposedly, low work function materials result in higher current densities relative to those materials with high work functions, as suggested by the widely applied Fowler-Nordheim theory. As a result of this widely, and perhaps incorrectly adopted theory, many have strived to develop low work function materials, composites, or coatings for enhanced field emitters. [16-18] [18] [17] Confirmation of the relative merits of low work function is, however, lacking. In this work, the effect of the material is studied in the largest meta-analysis of its type.

A considerable amount of data is available from a broad range of materials, which have been considered as viable candidates for field emission. No one, to date, however, has attempted to make a direct comparison between said materials. Only pure materials are considered herein; all adlayers and materials with surface coatings have been intentionally excluded from the study for simplicity. Ease of comparison between a range of materials allows for a comprehensive understanding of which materials are most suited for the use in various field emission applications. Differing field emission applications call for widely disparate electron emission performance, and a comparative knowledge of the available materials suited to said applications is technologically critical. In this paper we show for the first time, by considering the breadth of materials from published literature, that an electron emitter's work function does not have a significantly influential effect on the field emitting capabilities of a material, when the work function is the only comparative characteristic.

Here the studied materials are classified into categories according to dimensionality; namely, one dimensional (1D), two dimensional (2D) and three dimensional (3D)/bulk materials. The materials deposited or grown on the substrate, function as a field electron emitting cathode (**Figure 1a**). The liberated electrons tunnel through the restraining surface potential, whatever profile this may adopt, into ultra-high-vacuum conditions and are subsequently accelerated towards the anode. 1D materials are characterised by very high aspect ratios with nominal widths at the nanometre scale and typical lengths of at least one order of magnitude longer than their width. 1D emitters are diverse in structure, though often consist of aligned or disordered forests of 1D nanowires (NWs), which may be patterned, using conventional lithographic techniques, where the density of the 1D materials can be controlled either by the detailed growth conditions or the number of deposition cycles. 2D materials include the graphenes; a single sheet of hexagonally latticed carbon atoms, as well as the broader family of transition metal dichalcogenides. All are atomically thin, with typical single grains ranging in diameter from a few tens of nanometers, to many hundreds of micrometres. All 2D materials studied here were polycrystalline, and were either grown directly, or

deposited additively on various substrates, via processes much like the 1D nanomaterials. 3D and bulk nanomaterial emitters often have micro-cone geometry. They possess structures that consist of complex atomic and macroscopic arrangements; they can be crystalline, amorphous, disordered or structured. Nevertheless, their primary defining trait is their characteristically low aspect ratio, which is typically < 10. A number of materials in each of these categories have been used in field emission studies, with a particularly large number within the 1D set, attributed to an increase in interest in nanowires and nanotubes in recent history, and the sharp tips that they offer.

The key parameters for the success of a material as a field emitter are often depicted as a low turn on electric field, E_{on}, a low threshold electric field, E_{thr}, and a high maximum current density, J_{max}. Though key in assessing the emission performance, these metrics have been, to date, poorly defined, varying dramatically between publications [19-22], with many seemingly almost arbitrary definitions. Just under half of the papers studied herein give a value for E_{on} , usually with a value of 0.01 mA/cm^2. E_{thr} is stated less frequently, by approximately 20% of papers studied, at common values of 0.1 mA/cm^2, 10 mA/cm^2 and 1 mA/cm^2. Historically, the emission current density required to visualise electron emission patterns on phosphorescent screens is given as 10 nA/cm^2.[23] 10 mA/cm^2 is widely quoted as a “figure of merit”, particularly with regards to flat panel displays, though with no clear reasoning given as to why. [24] [25] The use of the commonly seen values in other field emission applications appears undefined, however, and they are not exclusively quoted. Indeed, some acknowledge that there are no strict rules, with some groups opting to define their own metrics.[26-29] Due to such arbitrary definitions and the apparent lack of consistency, it has proven, to date, prohibitively challenging to draw direct valid comparisons between differing materials and morphologies.

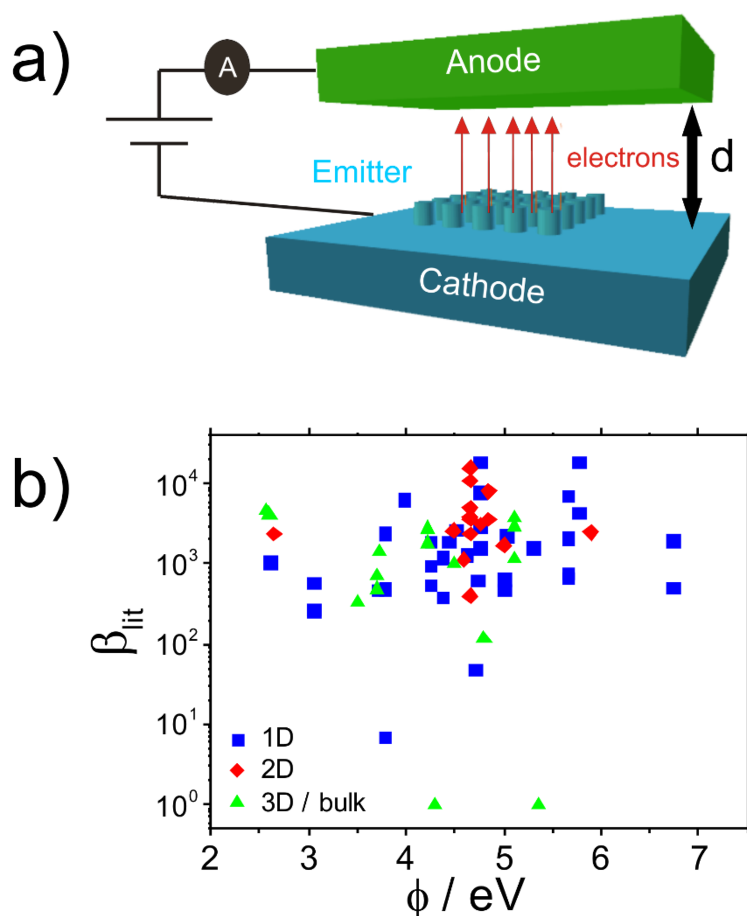

Figure 1 a) Generalised field emitting device. All studies considered herein use exclusively diode mode operation **b)** Emitter field enhancement factor (β) against work function (ϕ) for 1D, 2D and 3D/bulk materials, with little dependency seen.

In order to compare materials, a new definition was tested. **Figure 1(a)** shows a generalised field emitting device operated in diode mode. The emitting material, located in high vacuum conditions, is negatively biased and exposed to a high voltage, typically of the order of a few thousand volts on the anode. The inter-electrode vacuum gap (d)

defines the apparent global electric field. We generalised definitions for E_{on} and E_{thr}, defining them as 10% and 30%, respectively, of a normalised total measured current density, $J' = J_{max}/J$, when subjected to an applied electrostatic field (E). Data extracted in this way, across a breadth of studies, assuming consistency in J_{max}, can then be directly compared, allowing for the largest study to date across a near exhaustive range of materials. Data was extracted from 112 studies, previously published papers dated from 1984 to the present day. Where more than one paper per material was found, values of the extracted E_{on} and E_{thr} were averaged. Some materials, such as the carbon nanotubes (CNTs) and graphene, proved very popular, whereas with other, less popular materials, such as FePc [30] and cBN,[31] only a single paper was available. Work functions of the materials are given by an averaged value, where <n> ≥ 3. Having redefined the parameter E_{on}, to be 10% of the J_{max} value, however, some problems were highlighted. When J is normalised, the reading of 10% represents a different value of current density for each emitter that shows a different J_{max}. E_{on} directly relies on J_{max} for a value, therefore altering with overall performance. Whilst this has allowed a direct comparison to be made between materials, a similar comparison can be made by simply defining E_{on} as a single value of current density. We note that the two methodologies resulted in similar data sets, however we have opted for the latter approach given its simplicity and consistency with existing literature. The most commonly used value seen in the literature was 0.01 mA/cm^2, thusly justifying this otherwise arbitrarily chosen value. Not all materials could be measured, this was due to the range of measurements made individually, however, this was only around 2% of the studies considered.

In almost all studies to date, the measured FE properties have been mostly well-fitted with conventional Fowler-Nordheim tunnelling, as given by: $J = \left(\frac{A\beta^2E^2}{\varphi}\right)\exp\left[\frac{-B\varphi^{\frac{3}{2}}}{E}\right]$ where J is the current density, $A = 1.54\times10^{-6}$ A eV V^{-2} is a constant, ϕ is the emitter work function, B = 6.83 V $eV^{3/2}$ cm^{-1} is a constant, and E is the applied electric field. E can be approximated using the anode-cathode voltage (V) and inter-electrode separation (d) by $E = \beta(V/d)$, where β is the local field enhancement factor. The validity of the Fowler-Nordheim theory across most material platforms is certainly questionable, especially for materials that are not classical bulk metals. [32] Nevertheless, the emission current dependence on the materials work function has been widely implicated in various tunnelling models, as has the aspect ratio, or degree of perturbation in the emitting material. Nonetheless, the degree of suitability of models, such as Fowler-Nordheim, for materials where the tip has a radius of curvature the size of a single atom to a few nanometers [33] is still yet to be determined with any great accuracy. It can be seen that J, and hence J_{max}, can be tuned by augmenting ϕ and β, both of which can be altered by the surface geometry and chemistry. According to the general WKB approximation and subsequent transmission models, low ϕ and high β typically manifest as high J_{max}. In practice, however, for non-classical materials, such as nanowires and nanotubes, the intimate mechanisms which augment the emission are not yet fully understood. [34] This study focuses on the effect of changing ϕ across diverse material range in an attempt to rationalise the importance of emitter work function in comparison to the degree of perturbation in emitter geometries.

The extracted performance metrics (E_{on}, J_{max}) are organised according to the work function (ϕ), from lowest to highest. Another factor that is commonly implicated in affecting the field emitting performance of a material is the field enhancement factor (β). β relates the local electric field surrounding the emitter apex (E_0) to the linearly approximated macroscopic electric field (E), where $\beta = \frac{E_0}{E}$. Around 70% of the papers studied give β values, highlighting another inconsistency in the field. β is poorly defined, with some quoting it as the value of height (h) of the emitter over the radius of curvature (r) of the tip: (h/r),[35, 36] or some linearly scaled variation of this , with this scalar varying between 1 [27, 37-39] and 25 [28]. Others, more commonly (as is the case for all the 1D materials studied herein, and over 50% for 2D and 3D) state a value of β calculated by extracting it from a selected gradient on their coarsely fitted Fowler-Nordheim data. Some (7.5% of all papers studied) provide an empirical validation of such values by comparing them with β estimates using other methods, such as morphology estimates using electron microscopy imagery.[28, 38, 40] Others (2.5% of all papers studied) simply quote a value and suggest that β is a result of a combination of the emitter geometry; such as aspect ratio, surface roughness, the size of the vacuum gaps, crystal structure and spatial distribution of emitters. [41-43] It is not known, nor is any attempt made herein, to understand in

these cases, how each of these contributors affects β or indeed the emission properties in any great detail. For clarity **Table 1 (Supplementary Material),** shows an exhaustive list of definitions of β throughout the literature.

Whether there is a relationship between ϕ and β requires study. **Figure 1(b)** suggests that β from the literature, herein termed β_{lit} , does not appear to be a function of ϕ across the range of materials studied. **Figure 1(b)** highlights that the qualities most desired and strived for, and hence most commonly reported are low ϕ and high β, where a significant proportion of the data points lie at the top, with high β, and to the left of the figure, with ϕ < 5 eV . 1D materials show the largest spread in ϕ, whereas 2D materials are mostly confined to 4.0-5.0 eV, as they are at present predominately limited to the graphenes or other carbon based materials, as likely expected. 3D materials, on average, show a lower ϕ, reflecting the more traditional nature of these materials, but also a lower β than both 1D and 2D. A clear relationship cannot be seen between ϕ and β, although it is possible that ϕ can directly affect β (and vice versa), where ϕ (β) values are used to define β (ϕ) using the Fowler-Nordheim slope method: $\beta = -\left(\frac{b\phi^{\frac{3}{2}}}{k}\right)$ (**Table 1 – supplementary material).**

Ordering the extracted E_{on} and J_{max} performance metrics with increasing ϕ highlights the dependency of the material properties on field emission performance. **Figure 2** compares materials ordered by ϕ only, with no consideration to β or the surface morphology of the emitter. For each material the standard errors ($\langle n \rangle \simeq 3$, extracted from literature) are shown. The materials considered include the 1D nanowires - BaO [44] , LaB_6 [45] , Alq3 [40] [46, 47] [47], Si [37, 48, 49] [49], MgO [50, 51] [51], AlN [52, 53] [53], CdS [20, 54, 55] [55] , SiC [56, 57] [57], W [58, 59] [59], ITO [60], CuPC [61] , B [62, 63] [63], PPy [64-66] [65] [66], InGaN [28, 67, 68] [68], CNTs [21, 69-73] [70] [71] [72] [73], Cu [19, 74, 75] [75], ZnSe [76], diamond [77], GaN [78], ZnO [37, 79-83] [80] [81], ZnMgO [84], WO [85, 86], MoO_2 [87, 88] [88], and ZnS [37, 42] -, the 2D platelets – CuO [89, 90] [90], h-BN [91-94] [91] [94] [93], CbO [95], MoS_2 [96, 97] [97], graphene (monolayer, vertically standing, clustered and few layer) [22, 98-103] [99] [101] [102] [103] , RGO [100, 104], C nanowall [105, 106], WS_2-RGO [107], ZnO [108] and SnS_2 [109-111] [111] [110]-, and the 3D/bulk materials - *a*-diamond[82, 112], LaB_6[36, 39, 43, 113], nanodiamond [114, 115] [115], DLC [38, 116], *a*-C [29, 117], AlN [118], *ta*-C [119-122] [120] [121] [122], Si tips [35, 123], ZnSe [76], diamond [124, 125] [125], Cu tips [74, 75, 126], ZnO nanotrees [83], Ni tips [127-129] [129], chemical vapour deposition (CVD) diamond [130-132], and cBN [31, 133].

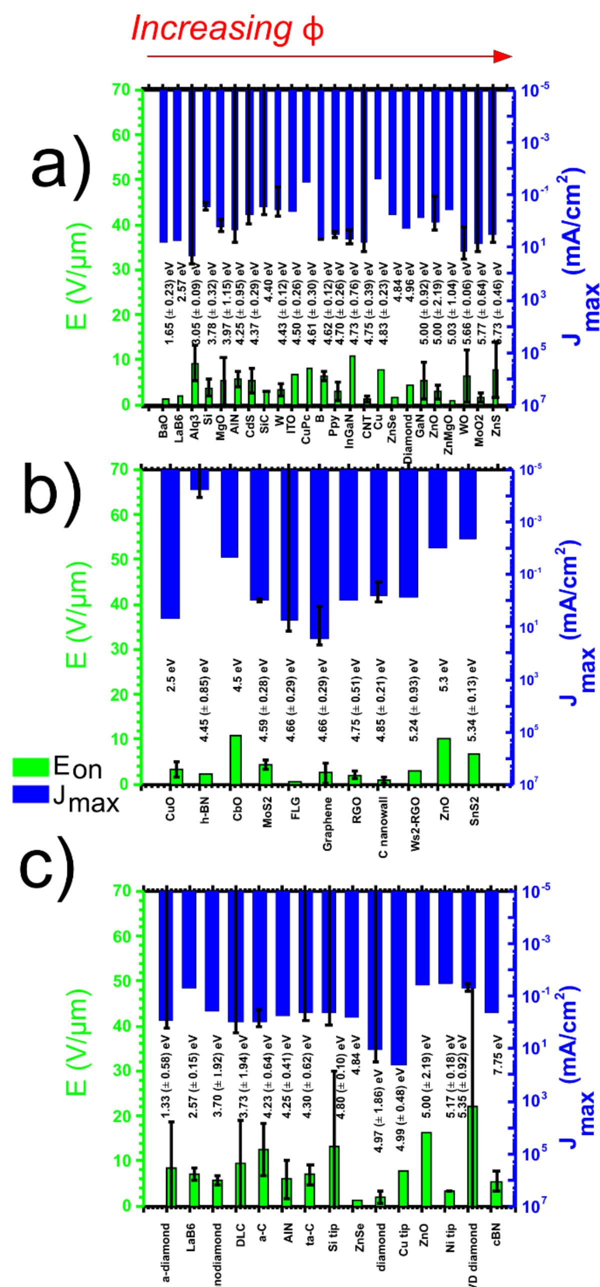

Figure 2. E_{on} (green) and J_{max} (blue) for **a)** 1D, **b)** 2D and **c)** 3D/bulk materials ordered by increasing work function (written above material). No distinctive trends were noted, suggesting that work function (ϕ) does not influence, to any great extent, a materials field emitting performance.

Comparisons can easily be made between materials in **Figure 2** when displayed in this way, both in regards to ϕ and on a material-to-material basis. It summarises a variety of field emission materials, considered across an intentionally diverse range of emitter geometries and morphologies in order to allow for a valid comparison of intrinsic material properties. When categorised by dimensionality it can be seen that the 2D and 1D materials have very similar average performance (1D: $\langle E_{on} \rangle$ | $\langle J_{max} \rangle$ = 4.74 V/μm | 3.61 mA/cm^2, 2D: 4.21 V/μm| 3.31 mA/cm^2). 3D/bulk materials, on average, require twice the electric field to achieve E_{on} as 1D and 2D (3D: $\langle E_{on} \rangle$ = 8.09 V/μm). This is likely due to the very sharp edges seen in 1D and 2D materials, with sizes down to the atomic range in some cases. A very similar average $\langle J_{max} \rangle$ (3D: $\langle J_{max} \rangle$ = 3.70 mA/cm^2) is seen across all materials, suggesting that this quantity does now strictly depend on dimensionality, and should be compared on an individual basis.

The graphitic allotropes, including the carbon nanotubes and graphene, show promising performance. CNTs show a low value of E_{on} = 1.28 V/μm compared to the average for 1D, as well as high maximum emission current density of the order of 6.76 (±7.45) mA/cm^2. Similarly, E_{on} = 2.52 (±2.16) V/μm was recorded for graphene, with a high J_{max} = 26.7 (±24.9) mA/cm^2 compared to the average for this dimensionality. Some materials such as ZnMgO nanowires (1D) and ZnSe (bulk) show promising values, with low E_{on} of 0.78 V/μm and 1.40 V/μm, respectively, but exhibit a poor comparison to the average of their respective dimensionalities with regards to J_{max} (0.35 mA/cm^2 and 0.63

mA/cm^2). In contrast to this, Tris(8 hydroxyquinolinato)aluminium (Alq3) nanowires (1D) and WO nanowires (1D) show remarkably high J_{max}, where Alq3 NWs have a $<J_{max}>$ of 20.5 (± 22.9) mA/cm^2 and WO NWs show a promising value of 13.8 (±12. 3) mA/cm^2 compared to CNTs with 6.76 (±7.45) mA/cm^2 . However, they do not consistently perform this well across all metrics, exhibiting higher E_{on}, with Alq3 NWs showing 9.23 (±3.93) V/µm and WO NWs with 6.37 (±5.75) V/µm. 2D materials, in particular the graphenes, show largely similar performance to one another.

Some claimed values of J_{max} vary widely within one material; the data for CVD diamond (polycrystalline), [130] showed E_{on} = 4.42 V/µm, with J_{max} = 0.11 mA/cm^2 , whilst another (which was hydrogen doped)[131] evidenced E_{on} = 58.40 V/µm, and J_{max} = 0.01 mA/cm^2. This results in a standard deviation of (E_{on}) 32.4 V/µm, which is larger than the mean (21.9 V/µm). This shows that there are evidently other, more central factors affecting the field emission capabilities other than simply ϕ of that material. Certainly in the present case the evident doping may affect ϕ slightly though certainly this would not be to the extent evidenced. With increasing maturity, increased consistency, and increased availability of data, performance metrics collected in this fashion will, in the future, likely reduce this limitation and the values found therein to become more reliable with respect to data collected for our other function studies.

In the ϕ ordered materials, no trend is seen on an expected exponential fitting, showing no seeming correlation between ϕ and E_{on} or J_{max}. It is expected, however, that the available data will expand rapidly with the continually emerging and expanding field of nano vacuum electronics, and henceforth a more defined relationship may become apparent. As prior, this future support the notion that there is more to the field emission capabilities of a material than simply ϕ arguments alone, and that other material characteristics have a larger effect on the field emitting capabilities. In combination with other characteristics, such as emitter morphology, and its evident manifestation in β, it is plausible that a clearer trend may be noted. Ordering materials in a combined and weighted ranking, such as a combination of ϕ and β, in addition to other metrics yet to be identified, may show an improved correlation with the extracted data. In **Figure 2** little dependence can also be seen on J_{max} with respect to ϕ. This is likely exacerbated by sample-to-sample measurement issues, such as minor variations in the measurement systems, as well as the extent to which the voltages of the emitters are driven by different groups. Ongoing systematic studies are underway to investigate the effects of the surface morphology, linked to β, on field emission, and the performance metrics, from materials that can be patterned as desired.

There are some factors that are not taken into account that could affect the outcome of the field emission properties of the materials investigated. Whilst, in many reported cases, ϕ is a defined bulk characteristic, surface ϕ of a material can be readily tuned to maximise emission. In practice, surface ϕ is not strictly constant and depends critically on the ambient. [134, 135] [135] The surface ϕ is particularly sensitive to chemisorbed absorbates, with subsequent hysteretic field electron emission studies observed, [136] necessitating *in situ* residual gas analysis, which highlighting the desorption of various species. This may well impact on the results from a single material, where otherwise nominally equivalent emitters have been chemically treated differently. In addition, the large data set size will likely induce some statistical scatter. Chief amongst which is the length (or height in the case of 2D and bulk materials) of the emitters. This is unlikely, however, to dramatically affect the results, and heights are likely to be within a degree of magnitude of one another. Another, potentially more significant factor affecting the emission properties is the cathode fabrication method employed **(Supplementary Material).** Such issues may include crystalline damage caused by cleaning processes, such as ultrasonication, different surfactants used in the fabrication process giving variations in dispersion and surface ϕ, as well as experimental conditions such as vacuum, temperature, and pressure levels, driving conditions and applied electrostatic fields, all of which have not been considered herein. The inter-electrode spacing may also have an influence on the field emission; however our data suggests that this is largely negligible in the present study (**Figure S1 - Supplementary Material**). Just 50 % of authors give a value of the inter-electrode distance, d. From those that did, however, d had a modal value of 100 µm. 86 % of values are within one σ of the mean (209 µm), suggesting that the data is largely unaffected by variation in d.

Another factor requiring consideration is the fabrication method, which around two thirds of studies stipulate. Due to the extent of the materials used, the number of methods employed reaches over 15. There are some materials (such as the CNTs) that can be synthesised using a number of techniques, whilst other, often newer materials, in general have only a single fabrication method. There is a possibility that this variation, seen amongst those materials

that have a number of fabrication methods, results in minor differing field emission behaviour between otherwise equivalent materials though our data suggests this is largely negligible compared to other, more dominant, variations in material parameters. The most common synthesis / fabrication method, however, across all dimensionalities is chemical vapour deposition (CVD), including the plasma enhanced and microwave variants. The high numbers seen using CVD is due largely to the various carbon based materials, which can be grown with desirable features, including alignment and patterning. Similarly, it is possible that *in* or *ex situ* doping and subsequent variations in the electronic properties of the material occur when CVD and wet chemistry methods are coupled. Even if the fabrication methods are similar, factors such as material composition, lattice configuration and alignment could all be different and may well affect emission performance dramatically. Nonetheless, the breadth of the study herein was designed to reduce the implications of these varied issues, with the resultant body of evidence indeed supporting our conclusions. Independent studies from different research groups were assessed to form a fair representation of each material; the focus here on ϕ. Though challenging to unify an otherwise disparate field, we have nevertheless using the present large data set endeavoured to produce the most concise summary to date of all the field emission materials across the 1D, 2D and 3D geometries, consistently evidencing only a very weak dependence on ϕ.

CONCLUSIONS

In the present meta-analysis we have directly compared the performance of the widest range of field electron emission materials to date. It was found that ordering materials by increasing work function did not result in any clear trend in turn-on electric field or maximum current density, suggesting other factors must be taken into consideration when discussing the field emitting capabilities of a material. E_{on} was found to be twice the size for 3D and bulk materials compared to 1D and 2D materials, suggesting the morphology of the emitter may be significant in regards to determining characteristics effecting field emission. Observations that can be made on a material-to-material comparison basis show that few materials seem more promising than the nanocarbons.

Experimental Section

A digital extraction tool (GetData Graph Digitizer, Vs 2.26.0.20) was used to digitise and gather data from the source metadata using the emission current density, J, as a function of the applied electrostatic field, E, data sets. In some cases, where current or voltage were given instead of J or E along an axis, data was converted into the correct form on the condition that the total emitting area or the cathode-anode separation was disclosed. The vast majority of the data ($\simeq 95\%$), however, was directly extracted from a J-E curve. This data was re-plotted and normalised.

The J_{max} value represents the maximum current density shown on the graph provided. This may result in variation of definition, whether the tip was run until it failed, array fraction, or maximum current extraction. In most cases, the value is assumed to be represented by the maximum current extracted.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Supplementary Material

To highlight the inconsistencies in the literature, an exhaustive list of the employed β definitions is given in **Table 1**. Around 70% of the papers quote a β value, regardless of the method they employ to extract β. Table 1 shows the extent of the range used to calculate β. There is a further discontinuity between material types, with 1D and 2D emitters being the most likely to disclose β; over 80% of 1D and 2D papers give β, with a mere 45% stating β in 3D/bulk. 80% of authors stating β use the Fowler-Nordheim slope, whilst 10% used only other methods (shown in Table 1). The remaining number of papers failed to state a method, simply claiming a value of β.

Table 1: Common β definitions

β – definitions		Ref.	Geometry
$\beta = (l/r)$	l = emitter length r = emitter radius	[30] [38]	1D/3D
$\beta = (h/r)$	h = emitter height (or length) r = is radius of curvature of tip or radius at apex	[39] [36] [35]	3D
$\beta = \left(\frac{d}{kr_{tip}}\right)$	d = inter-electrode distance k = constant (= 5 for long thin geometry) r = radius of curvature of tip	[128]	1D
$\beta = 100\left(1 + \sqrt{\frac{h}{2r}}\right)$	Smith's model: h = height r = radius of the emitter	[28]	1D
$\beta = \beta_0\beta_s = \beta_0\left[1 - \exp\left(-\frac{cs}{h}\right)\right]$ $\beta_0 = 1.2\left(\frac{h}{r} + 2.15\right)^{0.9}$	β_0 = intrinsic field enhancement factor is a ratio of local and macroscopic fields (r is radius). β = overall field enhancement factor, β_s = screening factor s = wire spacing c = constant.	[49]	1D
$\beta = \frac{1}{\left(\frac{h}{d} + \frac{1}{\beta_0}\right)}$	β_0 = enhancement factor (independent of d, h and applied voltage)	[86]	1D
$\beta_0 = \left(\frac{h}{0.95\,r_0}\right)$	β_0 = geometric field enhancement factor h = height r_0 = average radius of tip	[130]	3D
$\beta \approx d^{(1-c)}$	d = inter-electrode distance c = constant (< 1).	[106]	3D
$\beta = -\left(\frac{b\phi^{\frac{3}{2}}}{k}\right)$	b = constant (=6.83 x 10^7 $eV^{3/2}$ V/cm) k = Select gradient of the Fowler-Nordheim plot	All 1D that calculate β	1D, 2D and 3D

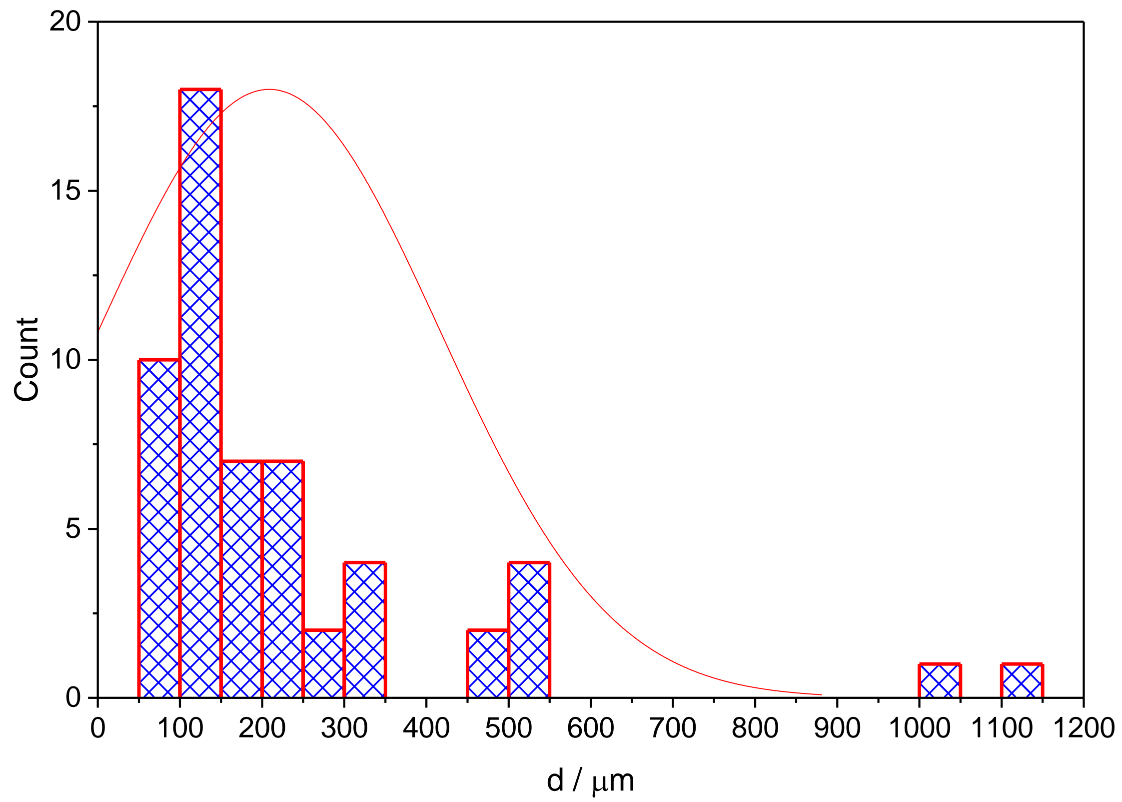

Figure S1. Histogram of the interelectrode spacing (d) of the considered studies.