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Review
Soft vacuum processing of materials with electron beams

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The use of soft vacuum (10^{-3} to 1 Torr) rather than high vacuum (10^{-5} to 10^{-4} Torr) in connection with electron beam processing of materials is discussed. It is argued that practical, technical and economic advantages can be realized in a wide range of applications from heat-treatment, through welding and melting processes, to vapour deposition and gas processing. The new technology of glow discharge electron beam sources is especially suited to soft vacuum processes. While mainly developed so far for welding applications, it can be applied to this much wider field which presently employs thermionic guns working in high vacuum conditions.

1. Introduction

Electron beam (E.B.) processing of materials began about seventy years ago when Pirani [1] applied a simple glow discharge gun to the melting of tantalum. However, it was not until about twenty years ago that powerful beams in high vacuum were applied industrially to melting [2] and welding [3]. Since then the technique has been commercially developed at a considerable pace and a number of profitable processes now depend on it [4]. Applications include heat-treatment, melting, refining, welding, machining and vapour deposition. Commercial welding guns are now available at powers up to about 25 kW and general purpose processing guns at powers up to 1200 kW [5] at beam efficiencies of about 70% [6].

These guns mainly depend on thermionic emission from a hot tungsten or tantalum cathode in high vacuum (10^{-4} to 10^{-5} Torr). They owe their feasibility to the engineering development of high vacuum for research in particle physics. High vacuum is a direct way to achieve adequately clean conditions in which to operate both processes and guns. It is now becoming evident, however, that many E.B. processing applications can be performed in soft vacuum. Thus E.B. welding in production line manufacturing processes is frequently carried out in soft vacuum [7] using differentially pumped high vacuum thermionic guns, with advantages in terms of pumpdown time and access time to the part being welded.

It is the aim of this paper to discuss the characteristics of soft vacuum and to show that it is possible to carry out heat-treatment, melting and evaporation processes in it with practical, technical and economic advantages. In particular, attention is also drawn to the suitability of soft vacuum electron guns of the glow discharge type [8], now in the stage of engineering development, for applications under these conditions. The paper therefore suggests that E.B. processing technology is about to turn full circle with a return to the methods first applied by Pirani.

2. Characteristics of soft vacuum

Soft vacuum for the present purpose means the gas pressure range 10^{-3} to 1 Torr. Electron beams penetrate a useful distance through gas in this pressure range [9, 10] (see Table I), the data being inversely proportional to pressure (strictly gas density). On the other hand, thermal atoms and molecules are transported by diffusion with relatively short mean free paths λ of the order of $10^{-2}/p$ cm, where p is the pressure in Torr. In a time interval t the molecules travel on the average a distance $r \approx 2D^{\frac{1}{2}}t^{\frac{1}{2}}$ in a random direction through the ambient gas, where $D \approx 1/3\lambda c$ is the diffusion coefficient and c the average speed. The m.f.p. for low energy ions

TABLE I Mean free path and range of electrons [9, 10] in air at pressure of 1 Torr

	Electron energy (keV)							
	1	2	5	10	20	50	100	200
M.f.p. (cm)	0.3	0.5	1.1	1.5	3.8	7.6	14	23
Range (cm)	6	16	52	200	800	3.8×10^3	10^4	3×10^4

(1 to 10 keV) is somewhat larger than for thermal molecules (e.g. by a factor ~ 3) and the range of energetic ions is of the order 2×10^{-4} E/p cm [11], where the energy E is in eV. Ions therefore penetrate less well than electrons.

In many material processing applications we are concerned with pumping the non-condensible gases, the transport and distribution of condensible vapours and the physical and chemical interaction of all gases with the material environment. Whenever an electron beam is applied to a surface some degree of gassing occurs and if the surface temperature rises sufficiently the material itself evaporates. In high vacuum molecules travel without collision to impinge upon the materials present. It is necessary to employ high volumetric pumping speeds to maintain a high vacuum and to use bent beam guns to minimize deleterious effects at the cathode. In soft vacuum, high mass pumping speeds are readily achieved and a deliberate throughput of a chosen ambient gas can be added to the gases and vapours liberated by processing. By proper design these process gases may be swept along by the ambient gas and back diffusion of contaminating vapours from pumps etc., may be avoided. The condensible vapour may be deposited in a desired manner at suitably disposed surfaces and the non-condensible gases can be pumped away rapidly through relatively small bore piping. The diffusion mechanism prevents condensible matter from penetrating far into ducts (not more than about 3 duct widths, or diameters) and so electron beams and other penetrating radiation can be brought into the processing region through ducts without the need for beam bending. Similarly, viewing ports looking through ducts straight at the process remain clean and uncontaminated by evaporated material and monitoring instruments can be operated without contamination problems. From the engineering and economic viewpoint soft vacuum systems have the advantage of fast pump-down and fast access for maintenance and other purposes.

Sometimes high vacuum is advocated on the grounds of purity of the processing environment. However, input control of the purity of the ambient gas in a soft vacuum can be readily applied to achieve the same, or even superior standards of purity. For example, inert gas is readily available pure to 1 part in 10^4 so that working at say a pressure of 10^{-1} Torr is then equivalent to a high vacuum of 10^{-5} Torr. In addition gas discharges may be readily set up in soft vacuum to clean, heat and degas all surfaces. If a hot processing chamber is employed (e.g. at a temperature of 200° C), thermal degassing and dissociation of organic contaminants quickly takes place. It is not always practical to have a hot main chamber and a more workable scheme in this respect is to employ a thermally isolated liner which may be heated deliberately or by the process itself. We have made tests on hot liners of this type used in vapour deposition processes. Even though multilayers of a variety of deposited materials are present on the liner, degassing rapidly occurs at 200° C or more such that purity of the atmosphere subsequently depends only on the quality of the gas input. It is not difficult by physical and chemical means to purify the input gas to the one part in 10^6 level in order to reach conditions of exceptionally high purity in the processing region. The choice of gas depends on the process to be operated. Obviously inert gas may be used for many processes, but essentially the gas must be compatible with the chemistry of the process and it is sometimes possible to choose other gases with advantage (e.g. hydrogen in the case of many heat-treatment, sintering, melting and vapour-deposition processes).

The writer has investigated the behaviour of condensible vapours in soft vacuum, particularly in relation to the electron beam evaporation of materials for coating applications [12]. In a cold chamber there is a strong tendency for precipitation to occur in the gas phase causing blacks or fine powders to be deposited on the various surfaces present. This effect has been noted by previous workers [13, 14] and has been developed by colleagues at Harwell [15] as a method of manufacturing extremely fine ceramic

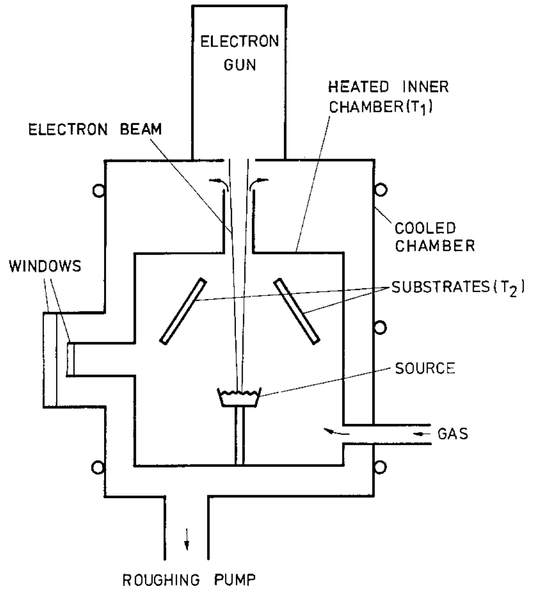

Figure 1 Soft vacuum E.B. deposition system (schematic).

powders (particle size is usually less than 10^{-8} nm). The theory of the mechanism of homogeneous nucleation responsible for this type of precipitation is understood only qualitatively at present [16-18]. However, inspection of the theory indicates that the process becomes less favourable as the gas temperature is raised [12]. It is possible in fact to find workable conditions in soft vacuum at gas temperatures of 200° C or more to avoid this effect. Transport of the vapour from source to substrate then occurs by molecular diffusion and dense heterogeneously nucleated deposits occur. Thus control of the chamber temperature in addition to soft vacuum pressure permits control over both the distribution and the physical characteristics of evaporated and deposited material.

The application of these principles to vapour deposition was demonstrated [19] with apparatus the main features of which are illustrated in Fig. 1. The inner chamber could be maintained at a temperature T_1 and the substrates at the same or a different temperature T_2. At an inner chamber temperature of 300° C dense, heterogeneously nucleated deposits were grown on the substrates (at the same temperature) at growth rates of the order of 1 μm min^{-1} at gas pressures of $400/d$ m Torr where d is the inner chamber dimension in cm. The electron beam power in the small experimental apparatus employed was about 500 W yet a wide range of ceramics and metals (including reactive and refractory metals) and a semi-conductor (silicon) were deposited in a pure state. The gases employed included both inert and reactive gases. A special case was hydrogen used for the deposition of aluminium, nickel, chromium, iron, stainless steel and molybdenum, and it was demonstrated that in this particular gas pressures as high as $4000/d$ gave satisfactory coatings. The adhesive strength depended on the chemistry of the situation and ranged from very high (e.g. most materials deposited onto glass substrates), to almost zero, (e.g. aluminium onto a nickel substrate). On the other hand, if the inner chamber and substrates were cooled, powdery deposits were made. It is interesting to note that, in a compromise situation of cold inner chamber but heated substrates, porous sintered fine-grain deposits could be obtained.

Further experiments at temperatures higher than 300° C showed that the ambient gas pressure could be increased significantly without inducing homogeneous nucleation (e.g. Ti was deposited at 600°C in argon at a pressure of $1000/d$ m Torr), and this is consistent with the theory [12]. It was found that the dense coatings made under these hot-gas conditions were put down with the high "throwing power" characteristics [20] to be expected from transport of molecular and atomic species by diffusion. Re-entrant angles, small cavities and even the backs of the substrates were coated. The structure of the coatings depended on the substrate temperature and it was possible to control the structure in the range amorphous through polycrystalline to single crystal, (e.g. sapphire was grown at a substrate temperature of 1400° C).

The inner chamber in this apparatus retains all the condensible vapour within it. None escapes through the vent which has a high aspect ratio (length/width). There are no seals used in the construction of the inner chamber which is assembled from component parts which fit together such that all the small gaps present have this high aspect ratio. The flow of ambient gas through the vent and also through these gaps prevents the back diffusion of oil vapours, etc. contained within the outer chamber. The windows remain uncontaminated.

3. Glow discharge E.B. sources

The principles on which these soft vacuum electron beam sources work has been fully described elsewhere [21]. The main features of a simple gun are shown in Fig. 2. A high voltage

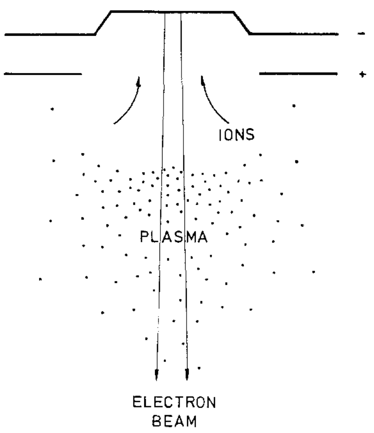

Figure 2 Main features of a simple glow discharge E.B. gun.

(1 to 100 kV or more) glow discharge takes place between the two electrodes in a soft vacuum in the pressure range 10^{-2} to 1 Torr depending on the specific design parameters. A plasma formed on the anodic side supplies energetic ions to bombard the cathode where they liberate electrons by secondary emission. The electrons are accelerated in the field between cathode and plasma to form an electron beam the shape of which is related to the specific electrode geometry. The diagram of Fig. 2 could represent a gun generating a thin pencil beam if the electrode structure has cylindrical symmetry about the beam axis, or the diagram could represent a section through a thin, sheet-like beam extending for a considerable distance perpendicular to the section. Again, rotation of the section about any axis lying in its plane would generate a structure giving a converging or diverging hollow beam (an extreme case produces a flat sheet-like beam converging on the perpendicular axis of symmetry). Other electrode configurations can produce a variety of characteristics and beam shapes, including broad, diffuse beams.

The choice of any particular gun design depends on the application. Simple and cheap guns can be employed for processes requiring continuous power densities up to about 10^3 W cm^{-2}. More sophisticated guns using magnetic focusing can achieve power densities of 10^6 W cm^{-2} or more, depending on the voltage of operation. In pulsed operation, high powers and power densities can be obtained even from simple guns. The beam power may be controlled either by regulating the gas pressure by means of a simple closed loop system, or by means of auxiliary electrodes. Beam efficiencies of 60 to 90% are obtained depending on design, characteristics and cathode material. In some cases where long life operation is required it is necessary to continuously rotate the cathode in order to minimize erosion caused by ion bombardment at the emitting region of the cathode surface. In some applications a different gas pressure or gas composition is required in the processing region from that in the gun. In these circumstances the beam is focused through a low gas conductance aperture or duct. The so-called point focus gun is ideal for this application, since owing to the high brightness [21] of the emission the axially symmetrical beam can be focused to a small diameter circle and it is possible to hold wide pressure differences (factor of 10 or more) across the aperture. The emerging beam can be refocused, or otherwise magnetically manipulated as required by the process.

Figs. 3, 4 and 5 show examples of practical guns engineered at Harwell for specific welding and other purposes. These guns work well in various gases, including air, at low voltages in the range 10 to 40 kV and at powers of several kW. High power (100 kW) and medium voltage (60 kV) guns are now under development and experimental work has indicated that high voltage guns, e.g. 150 kV or more, are feasible.

All these guns use robust cathodes which may be precisely engineered to give highly reproducible beam characteristics over long periods of time. The main problem in the design of glow discharge electron guns is to avoid arcing and flash-over within the gun. Our research has shown that these effects can be avoided if attention is given to two points: (1) the avoidance of volatile or easily evaporated material on the surface of the cathode, (2) the avoidance of gas pressure build-up between any part of the cathode assembly and surrounding metal at anode potential. These requirements are much easier to meet if the cathode and surroundings can be operated at elevated temperature, e.g. 200°C or more. Organic contaminants are then decomposed and driven off and sputtered cathode material is redeposited molecularly rather than as powder homogeneously nucleated in the gas phase. In addition, the hot gas in the gun environment brings the practical advantage of a higher working pressure since the discharge characteristics are essentially governed by the gas density. Thus once again the philosophy of a hot

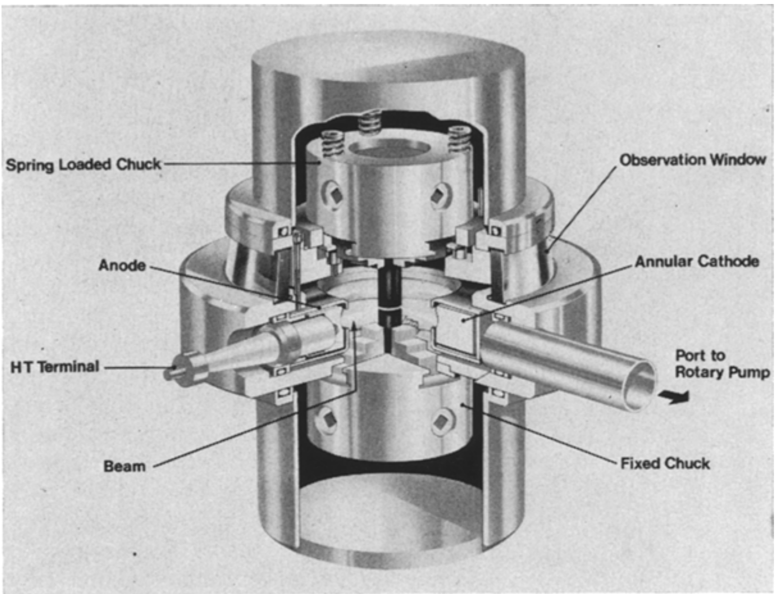

Figure 3 Ring focus gun for "single shot" tube welding.

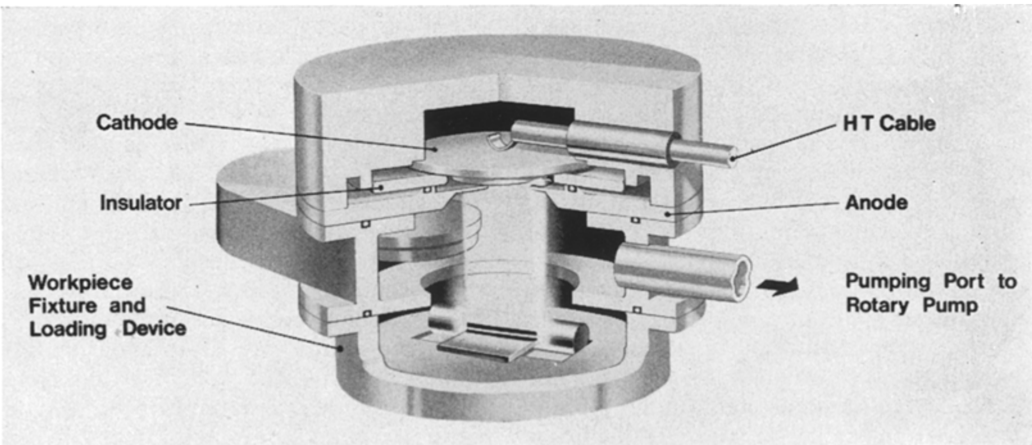

Figure 4 Line focus gun for "single shot" welding.

environment for the soft vacuum gives marked advantages in application.

We now have sufficient confidence to believe that very high power guns (hundreds of kilowatts) are entirely feasible and practical. Future development should aim to find cathode material with improved emission characteristics. The ideal cathode would have a high coefficient of secondary electron emission γ under ion bombardment and a low sputtering yield. It should work at high ambient temperature to both heat the surrounding gas and to dissipate the waste energy of ion bombardment by heat radiation to cooled surroundings at earth potential. In this way the engineering of high power guns could be simplified and the cathode losses (presently 10 to 20% of the power input using typical aluminium or stainless steel cathodes) could be reduced.

4. Applications

The main field of industrial application for glow discharge electron guns has been in welding and current developments in this field have been recently discussed [22]. Briefly, there are two areas, (i) the use of single-shot shaped beams

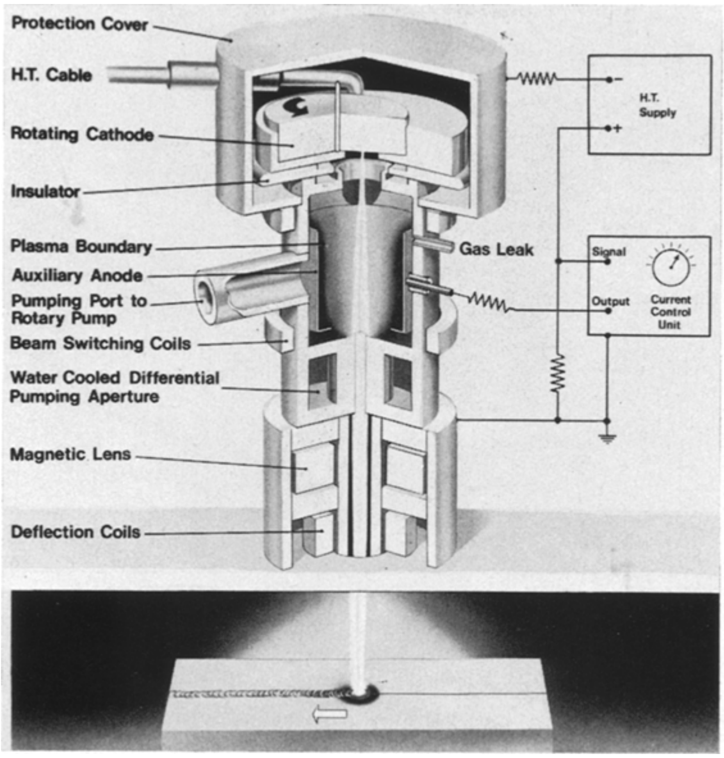

Figure 5 Point focus gun for general purpose welding and other applications.

which make the whole weld joint in one operation without movement of the parts (e.g. using guns of the types shown in Figs. 3 and 4), (ii) point-focus beams which make the weld joint by traversing in the conventional manner [23] (Fig. 5).

The single-shot shaped beams make a weld of the conduction limited type as in arc welding, but with a number of advantages (e.g. speed, absence of movement, purer atmosphere, uniformity and controllability and symmetry of thermally induced stress effects). The energy required in the shot is of the order of $2\ w^2l\ c\rho T_m$ where l is the weld length, w the thickness of the material, c the specific heat, ρ the density and T_m the melting point. The duration of the shot is of the order $w^2/\mathscr{K}$ where $\mathscr{K}$ is the thermal diffusivity of the material. A variety of metals and alloys can be joined in this way and the method has also been used to weld certain ceramics (e.g. silica, alumina).

Point focus guns can be sharply focused to weld thick materials and they can be designed to have a high degree of insensitivity to gassing from the workpiece. Guns of this type look promising for mass production welding operations. They can also be used for micro-machining, as electron probes and, owing to the ease with which they may be differentially pumped, to processing applications requiring controlled gas atmospheres at pressures governed only by the beam penetration characteristics.

A growing area of interest lies in continuous processing of wire and strip. There are three aspects, (i) heat-treatment for annealing, surface treatment, sintering, etc., (ii) radiation curing of organic coatings, (iii) coating by vapour deposition. This type of processing often demands that the wire or strip be fed continuously from a reel or magazine at atmospheric pressure through a set of differentially pumped seals into the processing chamber and similarly back to an atmospheric reservoir again. Considerable development of high vacuum technology to this end has already taken place [24]. These processes are all possible in a soft vacuum processing chamber, however, and as the glow discharge electron beam technology develops it should become economically attractive to simplify the production technique by cutting out the high vacuum stage. An experimental soft vacuum machine of this "air-to-air" type under develop-

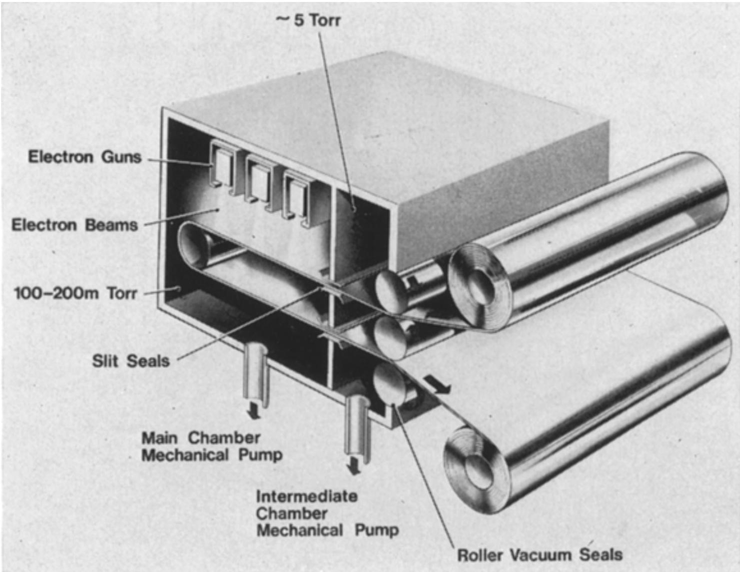

Figure 6 Plant for continuous processing of thin sheet (schematic).

ment at Harwell is shown in Fig. 6. With minor modifications the machine can be used for any of the above strip processing operations. Wire processing can in general be engineered more simply and we have constructed an experimental machine for the continuous coating of wire with either metals or ceramics (Fig. 7).

In a general way, soft vacuum vapour deposition seems to offer a wide range of applications for coating and forming. Many of the processes presently carried out in high vacuum [25] or by chemical vapour deposition [26] and other related techniques [27-29] can be performed by evaporation in soft vacuum using the principles discussed in Section 2. Deposition rates can be precisely calculated by the method of Szekely and Poveromo [30]. The applications range from thin films for electronic, magnetic and optical devices and for control over the mechanical and chemical properties of surfaces, through thicker coatings for corrosion, hardness and wear resistance, to forming of special components from intractable materials and composites and for crystal growing. Decorative coatings can also be applied in soft vacuum provided the process can be organized to meet the technical requirement for a hot ambient gaseous atmosphere. Not all processes are best done by heating with electron beams. Other forms of heat input, e.g. joule heating, radiation heating, induction heating, gas discharges, etc., may be economically more viable in some cases. Vapour can also be generated by sputtering and this method may be especially suited to the deposition of complex alloys and chemical compounds. However, whichever method of generating the vapour is employed, the essential feature of the soft vacuum process is the hot gas.

Special heat-treatment processes, for example, the melting, refining and casting of steel [31] and of refractory metal alloys [32, 33], could be readily carried out in a suitable inert soft vacuum using glow discharge electron guns. Volatile impurities can be swept away in the flowing gas and the condensation of evaporated metal can be confined within the processing chamber. Heat-treatments depending on the pulsing of surface temperatures to high transient values [34] can also be readily performed since it is possible to pulse shaped beam guns to high transient powers.

Finally, these soft vacuum guns can be employed to process gases themselves for chemical [35] and other purposes. A special application in this area is the ionisation of high pressure gas lasers for use in fusion plasma compression and heating [36, 37]. The glow discharge gun has the advantage, in relation to other electron sources, of simplicity, ruggedness and durability, and is capable of generating a broad beam at a high transient emission current density of the order of 10 A cm^{-2} [21] in

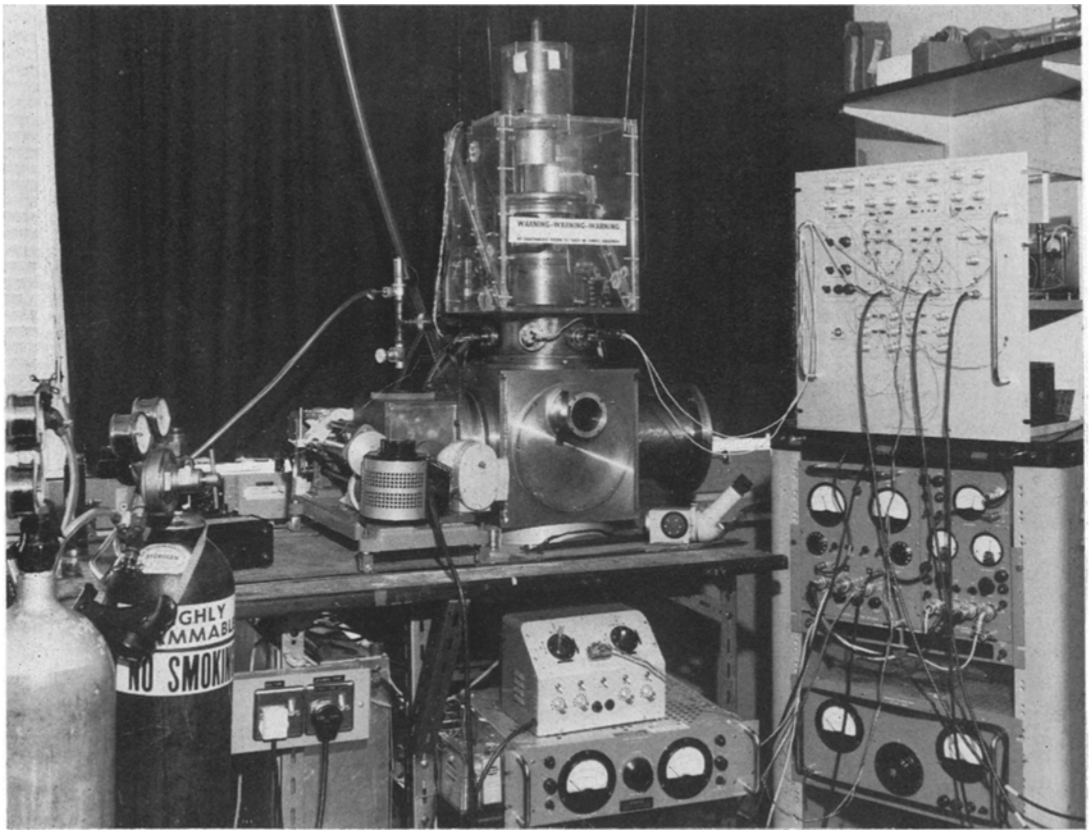

Figure 7 Experimental equipment for the continuous coating of wire (the supply and take up reels are enclosed in the extensions to the soft vacuum work chamber).

short pulses of a few microseconds duration.

5. Conclusions

The paper argues that many E.B. processes presently performed in high vacuum systems can be carried out in soft vacuum systems with practical, technical and economic advantages. Soft vacuum work chambers can be designed to give conditions of higher purity than are readily achieved in conventionally engineered high vacuum chambers. Volatile waste products are readily removed from the processing region and the distribution of condensible matter can be properly organized so as not to interfere with the means for processing and sensing. The current engineering development of glow discharge electron beam technology is, therefore, especially relevant since again only soft vacuum is required. While development so far has concentrated on welding applications there is every reason to expect that glow discharge guns can be engineered to high power and/or high voltage to match whatever requirements a particular process may demand. It seems just a question of time before the soft vacuum approach to electron beam processing largely displaces the present high vacuum technology.

Acknowledgements

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References

1. M. V. PIRANI, German Patent DP 188466 (February 1905).
2. H. R. SMITH, C. d'A. HUNT, C. W. HANKS, Proceedings of the 3rd Conference on Reactive Metals, Buffalo 1958 (AIME, Interscience, New York, 1958), p. 131.

3. J. A. STOHR, Fuel Elements Conference, Paris 1957. AEC-TID 7546 (Commissariat l' Energie Atomique, Paris, 1957) p. 9.
4. See, for example, R. M. SILVA (Ed.) Proceedings of the 2nd Electron Beam Processing Seminar, Frankfurt 1972 (Universal Technology Corporation, Dayton, Ohio, 1972).
5. S. SCHILLER, *ibid*, p. 1b1.
6. H. STEPHAN, *ibid*, p. 2a1.
7. *Anon*, "Electrical Beam Welding Review", Vol. 5 (Sciaky, Vitry-sur-Seine, 1974) p. 25.
8. R. A. DUGDALE, *J. Mater. Sci.* **1** (1966) 160.
9. B. W. SCHUMACHER, Proceedings of the 1st Electron and Ion Beam Science and Technology Conference, Toronto, 1964 (Wiley, New York, 1965) p. 5.
10. A. VON ENGEL, "Ionized Gases" (Clarendon Press, Oxford, 1955).
11. K. E. MANCHESTER, Proceedings of the 2nd Electron and Ion Beam Science and Technology Conference, New York, 1966 (AIME, New York, 1969), p. 1087.
12. R. A. DUGDALE, *Nature*, **249** (1974) 440.
13. L. HOLLAND, "Vacuum Deposition of Thin Films" (Chapman and Hall, London, 1956) p. 210.
14. O. REIFENSCHWEILER, *Rev. Sci. Instrum.* **35** (1964) 456.
15. R. G. AVERY and J. D. F. RAMSAY, *J. Colloid and Interface Sci.* **42** (1973) 597.
16. J. LOTHE and G. M. POUND, in "Nucleation" edited by A. C. Zettlemayer (Dekker, New York, 1969) p. 118.
17. J. D. MCGINTY, *J. Chem. Phys.* **55** (1971) 580.
18. A. G. SUTUGUN, *Phys. Letters* **29A** (1969) 233.
19. R. A. DUGDALE, in "Proceedings of 3rd Electron Beam Processing Seminar", edited by R. M. Silva, Stratford-upon-Avon 1974 (Universal Technology Corp, Dayton, Ohio, 1974) p. 2e1.
20. R. M. BURNS and W. W. BRADLEY, "Protective Coatings for Metals" 3rd Edn. (Reinhold, New York, 1967) p. 77.
21. R. A. DUGDALE, "Glow Discharge Material Processing" (Mills and Boon Monograph ME/5, London, 1971).
22. J. CONDLIFFE and R. A. DUGDALE, "Proceedings of the 14th International Machine Tool Design and Research Conference, Manchester 1973". (Macmillan, London, 1974) p. 611.
23. A. H. MELEKA, (Ed.) "Electron Beam Welding" (McGraw-Hill, London, 1971) p. 1.
24. H. R. SMITH, JUN, in [4], p. 1a1.
25. *Idem*, in [19] p. 2c1.
26. C. F. POWELL, J. H. OXLEY and J. M. BLOCHER, "Vapour Deposition" (Wiley, New York, 1966).
27. B. T. LUNDIN, (Ed.) "Proceedings of the Conference on Sputtering and Ion Plating", Lewis Research Centre, 1972 (NASA SP-5111).
28. R. G. KURTZ, "Vapour-Ion Methods of Film Deposition" (Dow Chemical Co. 1970, Report No. RFP-1568).
29. K. KELLER, *Härterei-tech. Mitt.* **26** (1971) 120.
30. J. SZEKELY and J. J. POVEROMO, *Met. Trans.* **5** (1974) 289.
31. T. H. HARRINGTON, *Canad. Met. Q.* **10** (1971) 137.
32. H. R. SMITH, JUN, "International Trans. Vac. Metallurgy Conference 1967" (American Vacuum Society, New York, 1968) p. 719.
33. C. MUELLER, in [4], p. 2d1.
34. See for example: Steigerwald Strahltechnik GMBH, German Patent DT 2209148 (September 1973), also British Steel Corporation, German Patent DT 2160805 (June, 1972).
35. H. G. FALSER, H. KOLLMAR and H. LUKAS, Battelle (Frankfurt) Information No. 11 (1971) p. 3.
36. A. CROCKER, H. FOSTER, H. M. LAMBERTON and J. H. HOLLIDAY, *Electronics Letters* **8** (1972) 461.
37. D. H. GILL, Los Alamos Scientific Laboratory Report LA UR 73224 (1973).

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