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Powder requirements for aerosol deposition of alumina films

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ABSTRACT

Dry powder aerosol deposition (AD) potentially offers several advantages over conventional ceramic coating processes. AD takes place at room temperature, and a variety of materials can be utilized as the substrate. Deposition occurs at a relatively rapid rate, and the product is a dense, nano-crystalline thick film. One limitation for AD is related to the suitability of powders. In addition to the many other process variables, the success of the deposition depends on the starting powder and its preparation, determined by trial and error. In this work, a broad experimental study about the suitability of 14 alumina powders for AD was carried out on two different substrates, alumina and glass. Three powder properties, particle size, d_{50}, specific surface area, S_{BET}, and compressibility index, *CI*, were measured and associated with the success of the subsequent deposition. For alumina substrates (high hardness), it was found that a relatively high powder specific surface area (5.5–8 m^2/g) correlated with good film deposition and adhesion. For glass substrates, initial deposition was obtained with almost all powders; however, problem with long term stability, apparently related to residual strain, was witnessed using many powders. Hi quality, well-adhered, stable AD films in this case were associated with comparatively lower S_{BET} and a compressibility index within a tight range of about 44–47%.

1. Introduction

The "Aerosol Deposition" (AD) method is a novel technique to deposit ceramic coatings with a wide range of thicknesses, from sub-micron to several hundred microns. Dense, nano-crystalline ceramic films result from the "room temperature impact consolidation" (RTIC) of dry powders [1]. High density is an outstanding property, usually exceeding 95% of the theoretical material density [2,3]. Any remaining pores in films are small, especially when compared to sintered bulk samples, with sizes between 15 and 100 nm [4,5]. These features distinguish the process from other types of additive manufacturing like aerosol jet spray [6], which is used to deposit relatively thin metallic or polymeric films, and requires a post chemical or thermal treatment. AD films can be prepared at room temperature without any heat treatment during or subsequent to the process. A rough vacuum is sufficient for a successful coating, reducing the effort, complexity and cost of process equipment [3]. AD can accommodate a broad spectrum of substrate materials, including metals [7,8], glass [9], ceramics [10], silicon [11], or even polymers [12]. Successful deposition has been shown for non-oxide materials (TiN [13], AlN [14,15], MgB_2 [16], MoSi/SiC [17]) as well as oxide materials (TiO_2 [18,19], ZrO_2 [20], Y_2O_3 [21], $Pb(Zr,Ti)O_3$ [22], $SrTi_{1-x}Fe_xO_3$ [10], BFT [23], $Bi_4V_2O_{11-\delta}$) [24]), and compound materials (Al_2O_3/PTFE [25], ZnS/diamond [26], Bi_2O_3/TiO_2 [27], Bi_2O_3/V_2O_5 [27]). Because of its wide applicability due to outstanding properties like high chemical inertness, wear resistance, and electrical resistivity, Al_2O_3 has been one of the most investigated ceramics [28–32], but still its behavior during AD is not well understood.

A basic AD apparatus consists of an aerosol generator, a deposition chamber and a vacuum pump (Fig. 1). Ceramic powders are kept in a continuously vibrating aerosol chamber. By passing a carrier gas through the bulk powder, a fluidized bed develops and particles are transported, driven by a pressure difference, from the aerosol chamber through tubes and a nozzle to the evacuated deposition chamber. The aerosol is accelerated by the nozzle to velocities above 150 m/s, forming an aerosol jet at the nozzle outlet. Powder particles collide with the substrate at high speed resulting in a breakdown of the starting particles to smaller fragments that form the ceramic film [3]. The film formation is believed to take place in two consecutive steps. In the first step, an anchoring layer (damage layer) is formed, resulting in a roughening of the substrate and the deposition of a monolayer of

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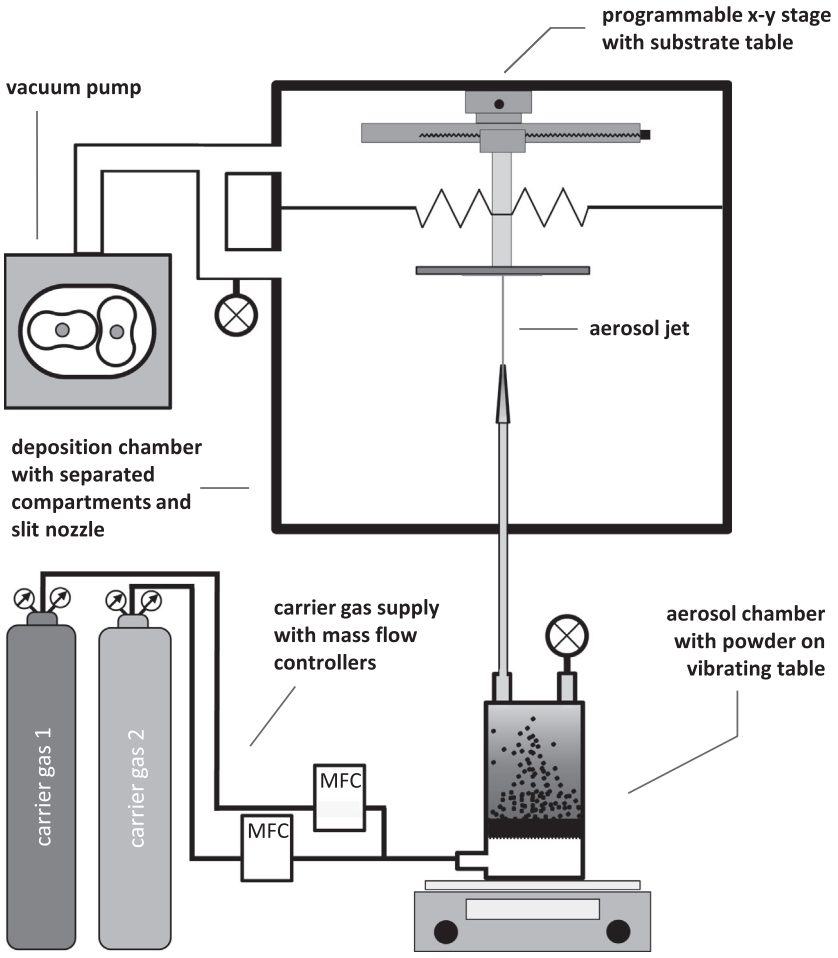

Fig. 1. Schematic drawing of an AD machine with aerosol generation unit, carrier gas supply, deposition chamber and vacuum pump. From Ref. [33] with the permission of the authors.

particles [22]. During the second stage, build-up of the film takes place by continuous particle impacts.

A major challenge for AD is the dependence of the resulting film quality on the initial powder and its preparation. Even when other process parameters are optimized, unsuitable powders only build chalk-like films [34], with the mechanical behavior of a green compact. Hence the right choice of raw powder is crucial for the success of the aerosol deposition. Powder characteristics are important at every stage of the operation, from generation of the aerosol, to particle acceleration in the nozzle, to impact, packing and bonding. Until now, only a very rough powder requirement is known, that is, a particle size between 80 nm and 2 μm [3]. But average particle size alone is not sufficient information, and is also complicated by agglomeration. Particle porosity, roughness, surface area, inter-particle friction, etc. are also expected to be of consequence. Compressibility index (*CI*), comparing bulk and tapped densities of a powder, is a simple measurement which is dependent on all of these. In conventional ceramic processing involving powder compaction, a low *CI* signifies a free-flowing powder, and is desirable. AD powders tend to be much finer, and the *CI* and its effects are unknown. The goal of this work was to identify such a powder parameter (or combination of parameters) which correlates with deposition behavior such that AD success might be better predicted. In order to accomplish this, a variety of Al_2O_3 powders were assessed using the 50% cumulative particle size, d_{50}, specific surface area, S_{BET}, and *CI*, along with the quality of resulting AD films.

2. Experimental

A wide range of powders with particles sizes (d_{50}) from 200 nm to 34 μm and specific surface areas (S_{BET}) between 0.3 and 10 m^2/g were chosen for investigation. This consisted of fourteen alumina powders from three sources (Almatis, Rotterdam, Netherlands; Baikowski, La Balme de Sillingy, France and Sasol, Johannesburg, South Africa). To minimize any chemical influences, all powders chosen were known (reported) to be primarily alpha-phase aluminas (no transition aluminas), and were of technical grade. Powders 1 through 11 (Table 1) are standard Bayer process aluminas, specified to have purity greater than 99%, with Na_2O content less than 0.1 wt.%. SiO_2 is typically the next major impurity, with content typically less than 0.03%. Powders 12–14 are described as high purity (>99.9%) aluminas, with major impurities Si and Na (approximately 20 ppm and 10 ppm respectively). This investigation was a study of the physical influences on deposition, and therefore details of the minor chemical differences were not considered further. Manufacturers' data (d_{50}, S_{BET}) of these powders are given in Table 1, marked "as-received". Powders were numbered and tested in random fashion so as not to introduce bias.

As a standard preparation for AD, each alumina powder was wet ground for 4 h in a high energy planetary ball mill [35]. Milling was conducted using zirconia media and cyclohexane as the liquid vehicle, followed by the removal of the solvent by a rotary evaporator. The powders were then sieved (mesh size 90 μm) to break down large soft agglomerates which negatively influence the