

THERMALLY SPRAYED COMPOSITE COATINGS OF ALUMINIUM AND SOL – GEL COATED SiC PARTICLES

Joaquin Rams, Belén Torres, Monica Campo, Alejandro Ureña
Universidad Rey Juan Carlos, ESCET, Dept. of Materials Science and Engineering
C/ Tulipán s/n, Móstoles E-28933 Madrid, Spain

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Abstract

Composite materials have been fabricated using thermal spraying from mixtures of powders of aluminum and SiC particles. Composites with reinforcement rates as high as 50 vol% were obtained with porosities of about 1% using a low pressure thermal spraying technique. This low porosity rates have been obtained because SiC particles were coated with silica following a sol – gel route. This coating is partially porous and improves the wettability of the SiC particles while avoids the appearance of aluminum carbide, a compound that degrades the composite.

The composites have been obtained as coatings and as bulk material. As a coating, it provides wear and corrosion resistance, while as bulk material it is a suitable material for electronics packaging. This fabrication process can be considered as a net shape technique.

1 Introduction

Surface of materials is in many cases the responsible of a product lifetime, in particular when corrosion or wear are involved. Coatings are a competitive method to improve their response. There are many coatings methods; among them, thermal spray is an interesting technology as it allows producing coatings up to several mm thick, and makes possible to deposit ceramics, polymers and metals as well as composite materials [1].

Substrates of aluminum alloys, and carbon steels have been used in this work. Aluminum alloys are lightweight, but their wear resistance is limited. Carbon steels show high elastic modulus and hardness, but they have a poor corrosion resistance. To improve the behavior of these materials, several coatings have been used in the literature, but many lack of combined wear – corrosion resistances, and other are not environmentally friendly. The use of

Al-SiC composites with high percentage of ceramic phase seems to be a good alternative to them.

The main problem related with these systems is the low wettability of SiC particles by molten aluminum at low temperatures and the formation of aluminum carbide at higher ones. We have used a sol – gel silica coating on particles that increases the wettability and avoids the formation of aluminum carbide [2, 3].

To fabricate composite Al – SiC coatings or bulk material, some authors have used plasma spraying or high velocity oxi-fuel, using milled composites as the feeding material in the spray gun [4, 5]. At a difference from these works, in this one, high quality SiC particle reinforced aluminum composite coatings have been obtained with a conventional thermal spraying procedure. Coatings were produced from mixtures of aluminum powder and sol-gel coated SiC particles, avoiding the need of casting the composite material powder and pulverizing it to appropriate mesh.

The composition, homogeneity and microstructure of the coatings are analyzed in coatings with different sol-gel treatments using scanning electron microscopy (SEM) and X-ray diffraction (XRD). The wear and corrosion resistance of coated samples have been also characterized on aluminum and steel substrates.

2 Experimental

The materials used to form the matrices were pure aluminum and an aluminum alloy containing 11% Si. In order to spray a composite coating, the metallic powders were mixed with 20 to 85%vol of SiC particles (SiCp) with 26 μm and 52 μm mean size. Particles were, in some cases surface treated with sol-gel silica coatings to improve the thermal spraying processing. Feedstock powders were prepared by a conventional rotating ball milling machine and afterwards the mixtures were directly used in a thermal spray gun. The substrates used

were 6082-T4 aluminum alloy and F-112 carbon steel blanks.

The sol – gel coating was prepared from tetraethylorthosilicate (TEOS) diluted in ethanol 1:11 and water 1:5. This mixture hydrolyzed for 2 h at room temperature under acid conditions. Particles were then placed in the sol and stirred for two hours. Afterwards, they were filtered, cleaned and dried for 1 h at 120 °C to evaporate water and constrained ethanol.

To consolidate the silica coating, samples were heated at 500 °C or at 725 °C for 1 h more to evaporate organic compounds and reduce porosity. The temperature used in this stage determines the specific surface of the coatings and, therefore, their chemical reactivity. In the case of 500 °C treated coating the specific surface, as measured by BET is 56 m²/g while in the 725 °C treated one this value lowers to 30 m²/g. It has been demonstrated that this difference in porosity modifies the interaction between molten aluminum and coated particles; the most porous coating shows a higher reactivity with the molten metal that gives higher wettability, while the 725 °C treated one is more passive and resists longer expositions to molten aluminum.

A low velocity oxy-acetylene thermal spray gun from Castolin DS8000 was used with a spray distance of 20 cm, neutral flame and a relative speed between gun and substrate of 150 cm/min. To enhance the adherence of the coating, substrates were grinded with corundum and were preheated at 200 °C. The thicknesses of the coatings obtained were in the 0.8 to 2 mm range.

To fabricate standing alone composites carbon steel substrates were used. To diminish the adherence of the sprayed material to the substrate, these were not heat nor were blasted with corundum. The oxide formed on the surface of substrates allowed easy removing of the coating. To reduce further the presence of pores in this material, the standing alone composites were hot pressed with a 2.5 MPa load for 30 to 60 minutes at temperatures in the 350 to 400 °C.

The Coated carbon steel substrates were tested in simulated sea water to determine the corrosion protection provided by the coating.

3. Results and discussion

3.1 Coatings on aluminum substrates

The fabrication of composites by thermal spray with uncoated particles on aluminum substrates

results in poor consolidated coatings. The lack of integration of particles inside the composite coating produces high porosity (figure 1a). When sol – gel silica coatings are laid on SiC particles (s-g SiCp), they get wet by the molten aluminum and are integrated inside the composite (figure 1b). As a result of it, porosity strongly reduces, reinforcement percentages increase and, then, the coating acts as a composite material. This phenomenon is common with both tested sol – gel coatings. There are minor differences between them which can be appreciated in Table 1 where different results obtained for different particle size, sol – gel coating used and % in the powder feeder are indicated.

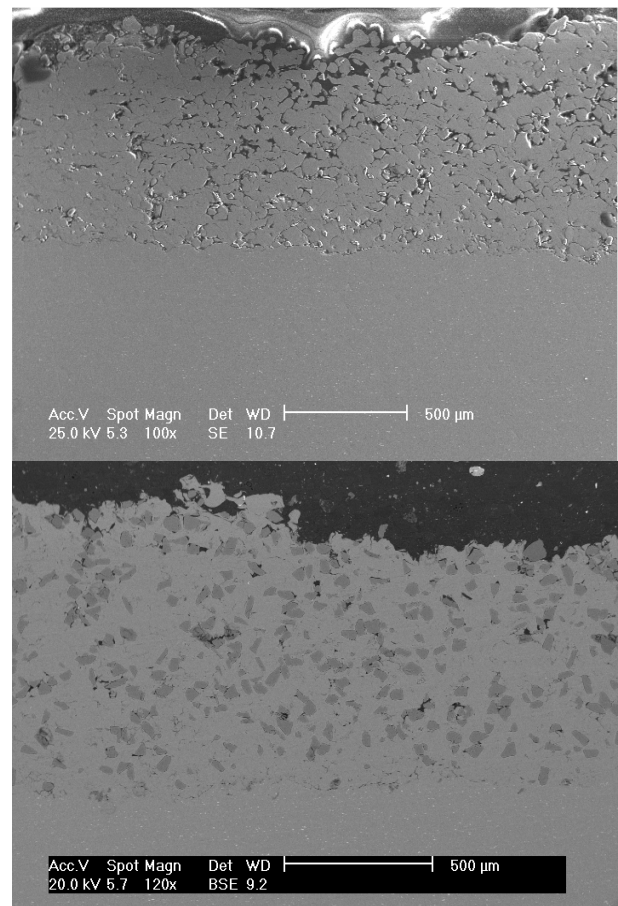


Fig. 1. Cross section of the sprayed coating on aluminum substrates: a) with SiCp b) with s-g SiCp.

Bigger particles give rise to better reinforced composites in all cases. The 500 °C treated sol – gel gives rise to less porous coatings, although minor differences appear between the two types of silica treatments, which are close to being considered as porous free composites. Surprisingly, in the case of s-g SiCp the porosity of the highest reinforced material is lower than that of the highest reinforced

one. This phenomenon is different in the case of uncoated particles in which porosity for mixtures of aluminum and SiCp of 50% or above cause the apparition of massive porosity. Figure 2a shows an image of this coating at a higher magnification. It can be seen that there is no real contact between reinforcement particles and matrix. This would result in the absence of load transfer between matrix and reinforcement, giving rise to low mechanical responses. The s-g SiCp are fully integrated in the matrix and aluminum surrounds them even when they have complex morphologies (figure 2b).

Table 1. Porosity and reinforcement rates in the sprayed coating for the different samples fabricated.

Size	% in feeder	Reinforcement vol% (porosity %)		
		uncoated	sol – gel 500 °C	sol – gel 725 °C
26 µm	20 vol%	3.0 (6)	4.1 (1.4)	4.6 (4.7)
52 µm	20 vol%	12.5 (2.3)	13.7 (0.9)	11.2 (1.4)
52 µm	50 vol%	18.1 (5.1)	27.1 (0.6)	24.2 (0.8)
52 µm	85 vol%	51 (6.0)	59.9 (0.5)	55 (0.7)

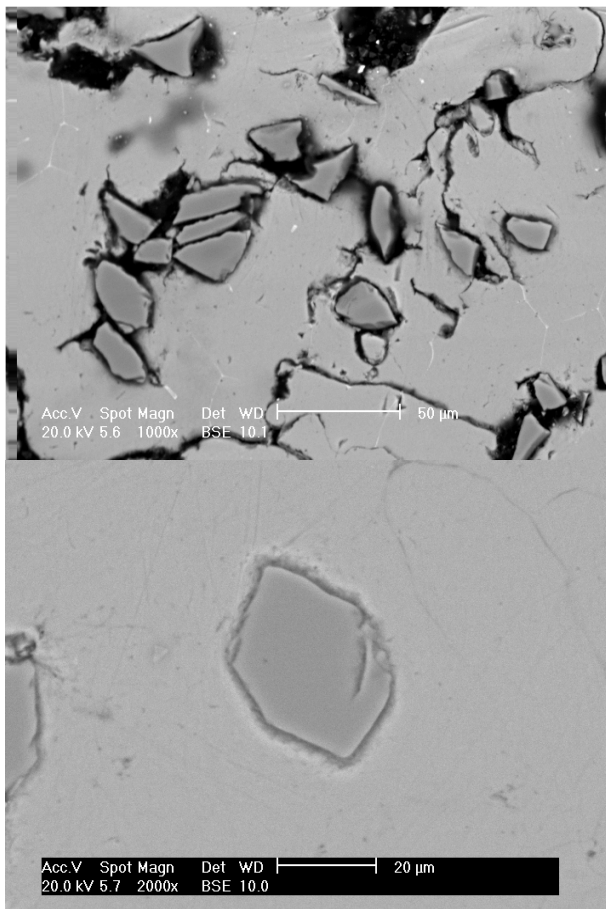


Fig. 2. Detail of SiCp: a) uncoated one and b) sol – gel coated particle.

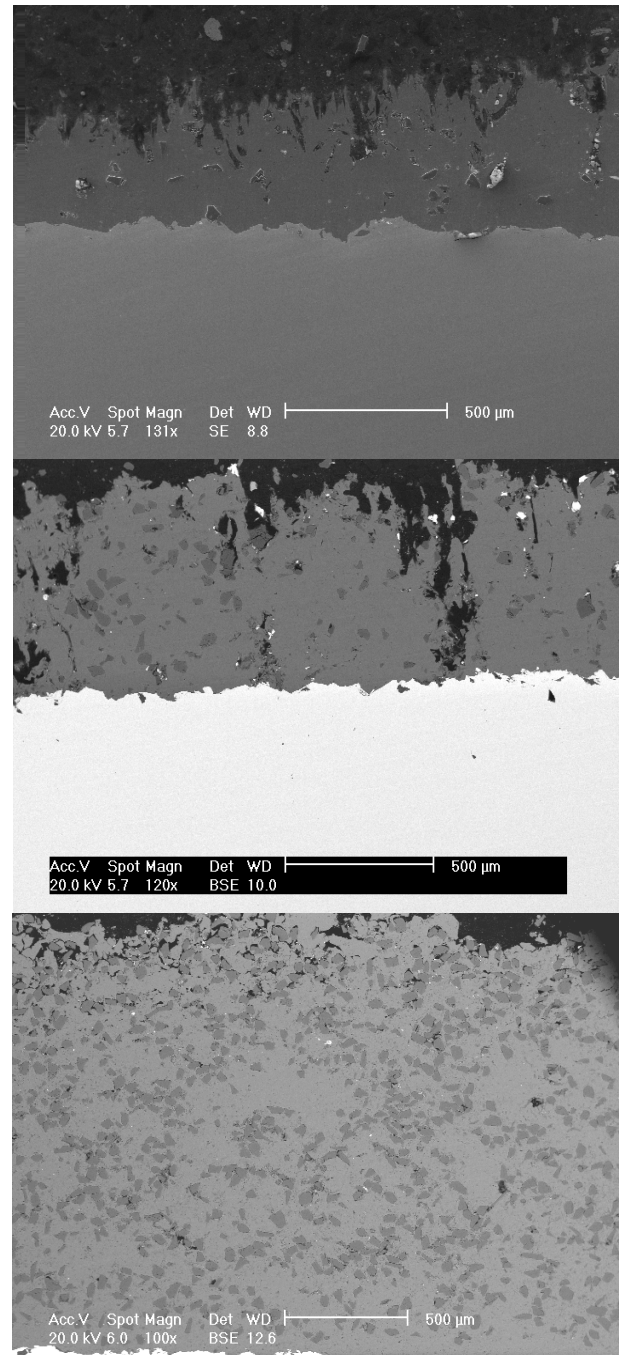


Fig. 3. Composite coatings on a steel substrate: a) and b) 20 vol% in feeder, and c) 50 % vol in feeder.

3.2 Coatings on steels

Most of the results observed in composite coatings on aluminum substrates reproduce when coating steel substrates. However, there is a peculiarity when coating steels with low percentage reinforcement (20 vol% in the feeder). Figure 3 shows the coating deposited on carbon steel used. The coating has broken in the transversal direction,

and some breakage lines even reach the steel substrate. This phenomenon can be explained by means of thermal stress in the coating that is originated from the different coefficients of thermal expansion (CTEs) of substrates ($12 \cdot 10^{-6} \text{ K}^{-1}$ for steel and $22 \cdot 10^{-6} \text{ K}^{-1}$ for aluminum).

To avoid the breaking of the coating we increased the percentage of particles in the feeding mixture and the result improved and correlate better with those observed on aluminum substrates (figure 3c). Reinforcement degree was above 25% and porosity was below 1%.

The main purpose of the coating laid on steels is to give protection against corrosion in aggressive media. We have made polarization tests to the coated materials and two main results may be observed. There is no contribution to the curve of the substrate, and in some cases there is an appreciable change in the corrosion potential that can be explained by means of the contribution of the sol – gel coating. In general, reductions of more than two orders of magnitude have been obtained in the corrosion sensitivity of the steel.

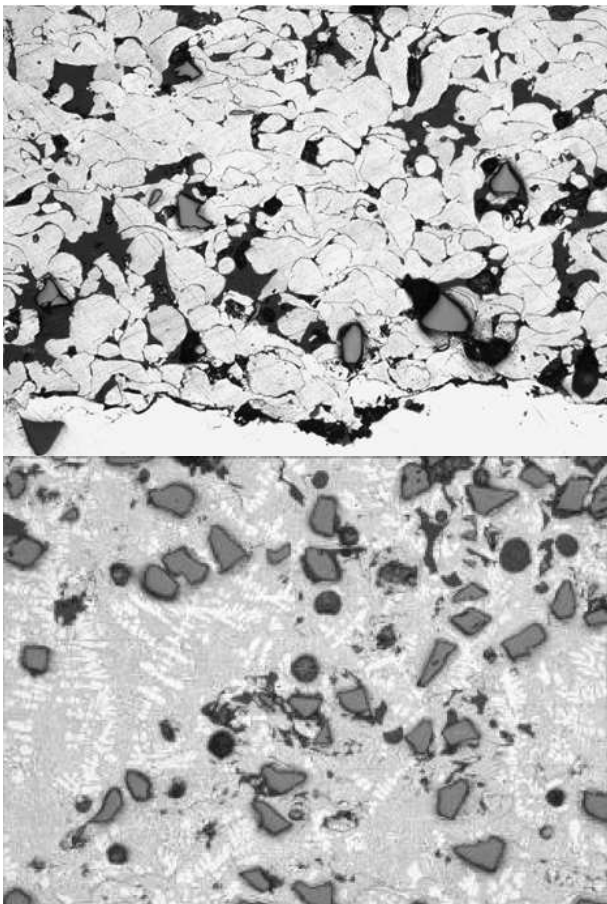


Fig. 4. Al-Si matrix composite with a) SiCp and b) s-g SiCp.

The pure aluminum matrix used is very useful for corrosion protection, but it is too soft to provide full wear protection to carbon steels. This has been solved using an Al-Si alloy. The wettability problems of the particles are increased by the presence of silicon in the matrix, and the coatings show big porosity when uncoated particles were used (figure 5a). It can be seen that the Al-Si droplets have not mixed altogether, and the coating is neither continuous nor compact. However, the incorporation of s-g SiCp allowed the wetting of the particles and a much better integrated coating was obtained (figure 5b) the continuity between the droplets that constituted the coating can be observed because dendrites appear on the matrix and the particle structure is not observable.

3.3 Fabrication of bulk composite materials

After composite coatings have been sprayed we decided to fabricate coatings that were not adhered to the substrate in order to remove it easily from it to fabricate a standing alone material with a 1 to 2 mm thickness.

The material was sprayed on a polished and oxidized steel sample to reduce the adherence. The substrate was also kept at room temperature before spraying for the same purpose.

The shape of the as-sprayed composites (figure 5a) is similar to that shown on coatings sprayed on steels but with a lower porosity. This is because of the little contact with the substrate used for spraying which limits the heat transference from the sprayed material of the coating to the substrate. Due to this, and considering that the mass of the coating is much lower than that of the substrate, the sprayed material reaches higher temperatures and aluminum is better molten, giving rise to a better integration with the particles. As in previous cases, the adoption of a sol-gel silica layer on the particles reduces the porosity of sprayed coatings.

After compressing the material with the hot plates for 30 minutes at $400 \text{ }^{\circ}\text{C}$ most of the porosity has disappeared (figures 5b and c), although some remains in localized zones. Most particles are fully integrated in the matrix and they are homogeneously distributed in the composite. In the case of the highest reinforced material the reinforcement rate reached 50% and aluminum was homogeneously filling all the spaces between the particles.

When observing the samples with more detail (figure 6), it can be observed that there is full

integration between particles and aluminum, and that aluminum carbide, i.e. the product of the direct reaction between aluminum and SiCp which degrades these type of composites, has not formed.

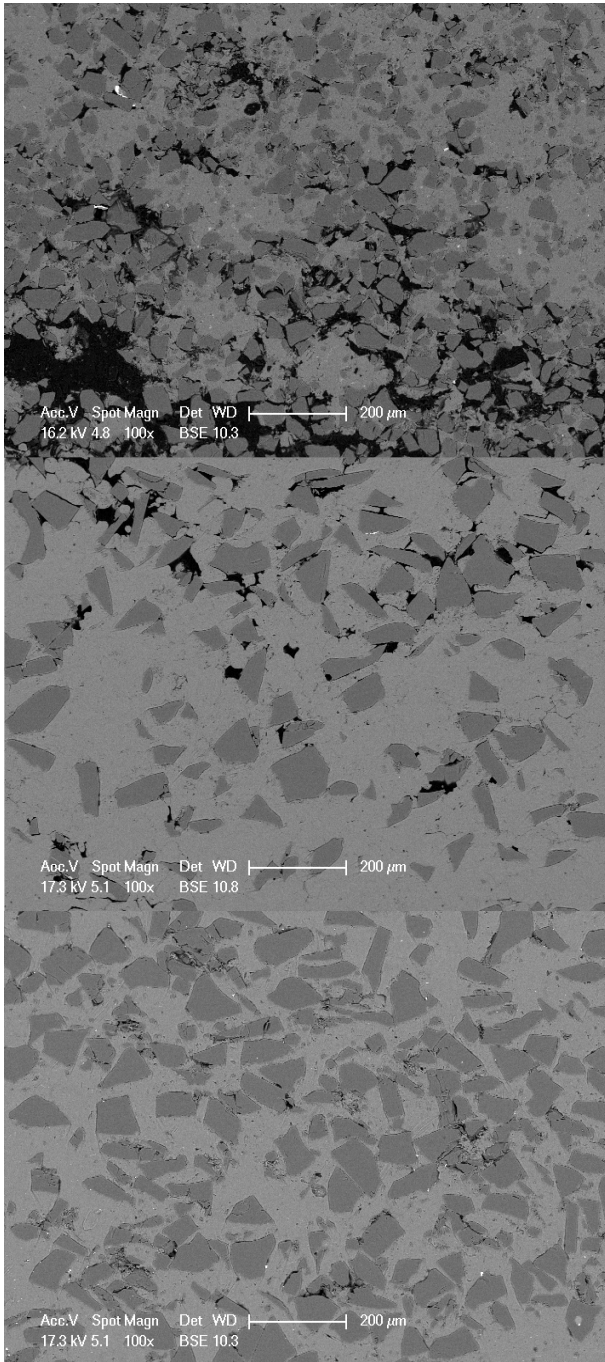


Fig. 5. Thermally sprayed composite: a) as-sprayed, b) hot pressed 50%, c) hot pressed 70%.

The process shown allows obtaining composites with high SiCp content from mixtures of treated or untreated SiCp and pure aluminum or aluminum alloys without the need of making a

perform. Particles are homogeneously distributed in the matrix and are perfectly integrated in it. The process shown can be considered as a net shape fabrication one because the thickness of the coating and its size can be easily controlled by the displacement of the heat to the thermal spraying equipment. The final measures of the sample can be controlled using the appropriate mold at the hot – plate machine. Finally, the surface finishing provided by hot plates and by thermal spraying, in its side facing to the substrate, is good enough for direct commercializing.

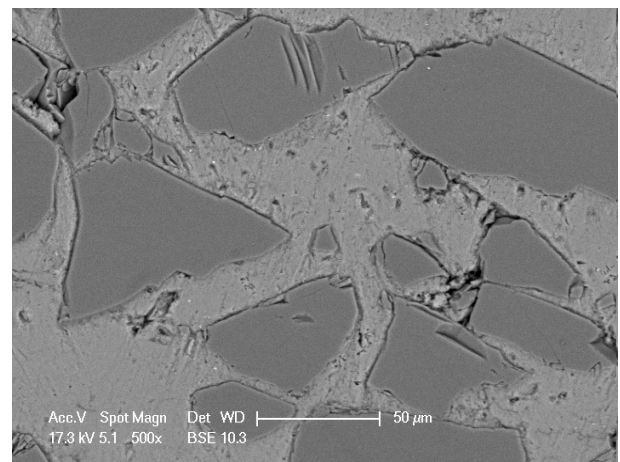


Fig. 6. Detail of particle – matrix interaction in thermally sprayed hot-pressed composite.

4 Conclusions

- Thick dense Al – SiC composite layers have been obtained through conventional thermal spray from a mixture of Al powder and sol – gel silica coated SiC particles on aluminum and on steel substrates.
- The use of a sol-gel interfacial layer allows reducing porosity and increasing reinforcement rate and load transfer between matrix and reinforcement.
- The wear resistances obtained in reinforced coating are up to 10 times higher than that of unreinforced coatings. On carbon steels, the coating acts as a corrosion resistant coating as well as a wear resistant one.
- Bulk composites were fabricated by means of thermal spraying. The material obtained had little porosity which was reduced by means of compressing the samples at temperatures above 300 °C.

References

- [1] Herman H, Sampath S, McCune R. “Thermal spray: Current status and future trends” *MRS Bull*, Vol. 25, No. 7, pp 17 - 25, 2000.
- [2] Rams J, Campo M, Ureña A. “Sol-gel coatings to improve processing of aluminium matrix SiC reinforced composite materials” *J Mater Res*. Vol. 19, No. 7, pp 2109 - 2116, 2004.
- [3] Rams J, Ureña A, Campo M. “Sol-gel coatings as active barriers to protect ceramic reinforcement in aluminium matrix composites” *Adv. Eng. Mat* Vol. 6, No. 1-2, pp 57 – 61, 2004.
- [4] Gui MC, Kang SB. “6061Al//Al-SiCp bi-layer composites produced by plasma-spraying process” *Mater Lett* Vol. 46, No. 5, pp 296 – 302, 2000.
- [5] Podlesak H, Schnick T, Pawlowski L, Steinhäuser S, Wielage B. “Microscopic study of Al-SiC particulate composites processed by laser shocks” *Surf Coat Technol* Vol. 1, No. 124, pp 32 – 38, 2000.