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HEAT RESISTANT ULTRA-FINE GRAINED Al PROFILES

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Abstract. Ultra-fine, monocrystalline, gas atomized Al 99.7% powder was consolidated into profiles by means of direct extrusion. Conventional approach, with complicated (i.e. expensive) procedure steps of powder degassing and encapsulating, was avoided. Structural stability of compacts and enhanced mechanical properties, predominantly those at elevated temperatures, were of paramount interest. Extrusion yielded sound profiles with ultra-fine grained (UFG) microstructures of fcc-Al matrix reinforced with homogenously redistributed α-alumina dispersions. Nano-scale dispersions, originated from torn surface oxide's envelopes, effectively acted as dislocation movement and grain growth barriers. Microstructures were found to be intact to long term high temperature exposures. Relatively high tensile strengths (R_m up to 310 MPa) accompanied with good ductility (A_{10} ~10%) and impact fracture toughness (KC ~45 Jcm^{-2}) were determined at room temperature. Due to unique structural stability no pronounced strength decline was observed and compacts retained their relatively high strength even at testing temperatures of 500 °C (~70 MPa). Simple consolidation route without prior degassing resulted in the structure containing micropores filled with entrapped and compressed gasses. These pores had no negative effect on room temperature mechanical properties, though expansion of entrapped gases may lead to the reduction of high temperature ductility.

1. INTRODUCTION

The large surface area of ultra fine Al powders gives a rise to possibility to introduce relatively high amount of naturally formed surface oxide into the structure of subsequent powder compacts. Thus metal matrix composites of Al matrix strengthened with nano-metric torn surface oxide dispersions with thickness of ~5 nm [1] can be produced in relatively easy way. It would be extremely technologically difficult to bring and redistribute evenly such a small dispersions into structure intentionally if done by other techniques. To break up surface oxide's envelopes, to maintain desired ultra-fine grained (UFG) compact's microstructures and to establish good metallurgical bonding between particles the consolidation is basically circumscribed to compaction routes based on introduction of shear deformation (mostly extrusion) [2].

Nano-metric dispersions do not tend to act as micro-concentrators, are not susceptible to cleavage and do not cause formation of voids [3]. As a result superior mechanical properties can be attained at substantially lower dispersion contents when compared to micro-metric size counterparts. Moreover oxide particles grain pin the structure yielding unique structural stability of compacts even during long term high temperature exposures. It was expected that proper consolidation of fine Al powders would result in profiles with superior mechanical properties, especially at elevated temperatures. This feature would be in contrast with sudden strength deterioration at even moderate tem-

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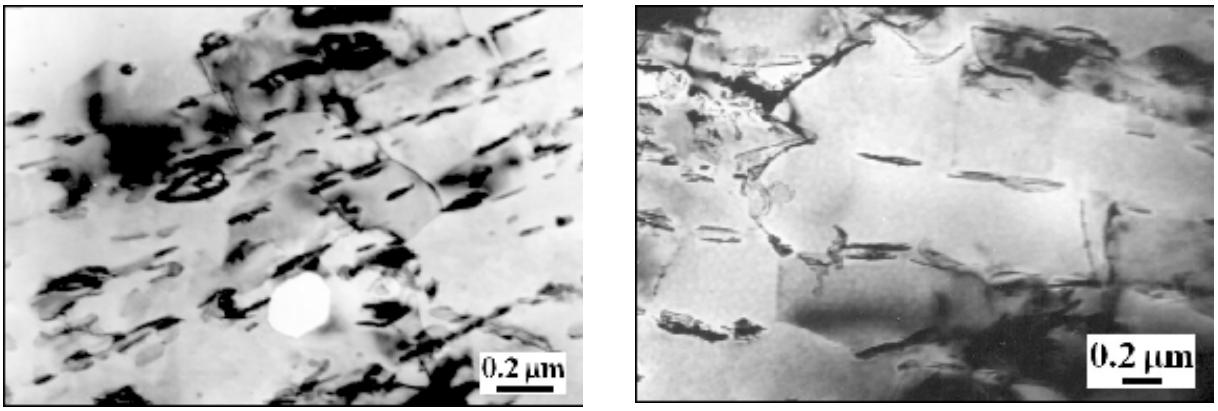

Fig. 1 TEM cross-sectional micrographs (BF) of 1 μm Al powder compacts extruded at 350 °C (left) and 500 °C (right) at *R*=11:1. Longitudinal to extrusion direction.

peratures of conventional high strength Al based alloys based on precipitation strengthening due to overaging processes.

In order to desorb oxygen, water and other oxidizing gases absorbed on the surface of powder, extrusion of encapsulated and degassed powder is widely used [1, 2, 4-7]. On the other hand procedure of hot degassing and encapsulating of particles prior consolidation considerably surcharges the price of final compacts. In order to keep the production expenses low and thus maintain potential industrial process feasible, these processes were intentionally avoided in this work. Although, the negative effect of entrapped gases was carefully investigated.

2. EXPERIMENTAL

Spherical, monocrystalline, 1 μm (d_{50}=1.31 μm) Al powder of technical purity 99.7% (O_2 content 2.05 wt.%) was prepared by gas atomization in N_2 atmosphere [8]. Coarser 10 ìm and 63-400 μm fraction Al 99.7% powders were used for comparison.

Powder precompacts were prepared via CIP at 200 MPa. Compaction was realized via conventional direct extrusion with flat faced die at different extrusion ratios (11:1, 20:1, 44:1). CIPed powders were filled into the preheated die and heated up to desired compaction temperatures T_{ext} for 30 min before consolidation. The average ram speed during extrusion was ~1 mm.s^{-1} (±0.8). BN spray was used as a lubricant. See complex study related to consolidation behaviour of ultra-fine Al powders during hot extrusion elsewhere [9].

Structures of extruded rods were examined by means of light microscopy, SEM and TEM. To clearly demonstrate presence and distribution of nano-scale oxide dispersions, compact's samples for TEM observations were electrolytically etched. Relative density of the extrudates was measured via Archimedes route. The mechanical properties at room and at 300 °C were measured on specimens with the gauge of Ø3–30 mm (tension) and Ø5.5-10 mm (compression) using ZWICK testing machine at the cross ram speed of 1 mm.min^{-1}. Vickers microhardness measurement were performed on compacts cross section with applied load of 10 g. Due to small dimension of final compacts, the unnotched specimens for impact Charpy fracture toughness measurement had non-standardized dimension 4x4x55 mm (machined longitudinally to the extrusion axis).

Dilatometry experiments of PM compacts were performed using thermal–mechanical analysis equipment TMA 2940 CE, Thermal Instruments (held under nitrogen) [10]. The vertically positioned 4 x 4 x 10 mm samples were heated up from room temperature up to 550 °C and cooled down to room temperature in three cycles at the rate of 3 °C·min^{-1}.

3. RESULTS AND DISCUSSION

3.1. Microstructural characterization of powder compacts

Relatively high densities in the range of 2.66-2.7 g·cm^{-3} were determined for extruded compacts of

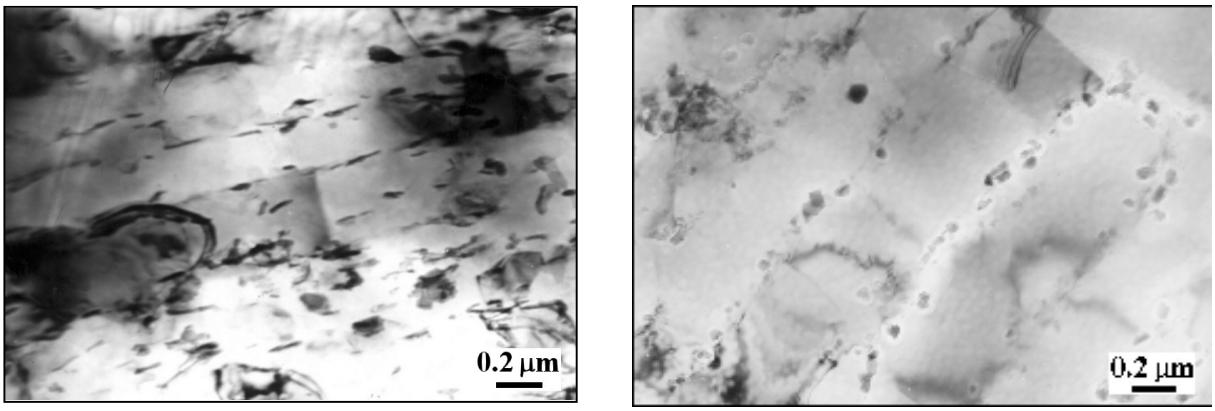

Fig. 2. TEM micrograph (BF) of 1 μm Al powder compacts extruded at 350 °C after annealing at 350 °C (left) and 500 °C (right) for 20 hours. Longitudinal to extrusion direction.

all experimental powders. The deviation from ideal aluminium density (2.7 g·cm^{-3}) is due to two competing phenomena: the presence of residual porosity and the small amount of Al_2O_3 oxide phase of higher density (3.66 g·cm^{-3}). Since the oxide thickness is independent of particle diameter and its content is proportional only to particles surface [1], oxide weight contribution in case of coarser 10 mm and 63-400 mm Al powder compacts was negligibly small.

All compacts extruded from 1 μm powder at extrusion ratio 11:1 exhibited approximately the same density (~ 2.67 g·cm^{-3}) regardless of extrusion temperature (T_{ext}). Increase of extrusion ratio (R) from 11:1 to 20:1 and 44:1 slightly improved density to 2.68 g·cm^{-3} and 2.69 g·cm^{-3}, respectively. Expectedly smaller surface area of both coarser powders led to smaller internal porosity of their corresponding compacts. In this case the density reached the values at the level of ~ 2.69 g·cm^{-3}, again regardless of the particular T_{ext}.

TEM micrographs of extruded bars revealed deformation and fragmentation of initial powder particles of all studied powders into elongated grains aligned along to the extrusion direction. As initially spherical powder particles elongated, surface area enlarged, what led to eventual oxide envelopes breakage. Diffusion at newly uncovered surface areas gave a rise to a formation of metallic bound “bridges” between individual powder particles.

Extensive energy imposed to 1 μm powder during extrusion at low temperatures (350 °C), see [4], resulted in heavily deformed powder particles, superior fragmentation of surface oxides (shred-like particles dark in contrast) and in homogeneous distribution of these fragments even inside of newly arose fine grains (Fig. 1). On the contrary extrusion held at T_{ext} = 500 °C did not ensured such fine microstructure as in case of T_{ext} = 350 °C and torn oxides predominantly decorated grain (particle) boundaries. Transversal grain size of extruded 1 μm powder compacts was determined to be ~200 nm.

Further increase of induced strain (i.e. higher R=20:1 and 44:1) did not led to finer microstructures, conversely a few recrystallized areas were observed if higher T_{ext} = 500 °C was used. This might be attributed to the limits in reachable work grain refinement and the start of dynamic recovery processes. Moreover higher amount of heat developed during extrusion at higher R might accelerate dynamic recrystallization.

Annealing experiments (350 °C@20 hours) proved the role of nano-scale dispersions in preventing excessive grain growth at elevated temperatures of 1 μm Al powder compacts. These experiments accommodated no major microstructural changes, where microstructure of annealed compacts retained unchanged (Fig. 2). Further shift of annealing temperature towards to the relatively extreme 500 °C@20 hours led to apparent grain growth (Fig. 2), although the structure remained still reasonable fine. Oxides dispersions were found only at the grain boundaries. Moreover due to surface area minimization they became spherical in

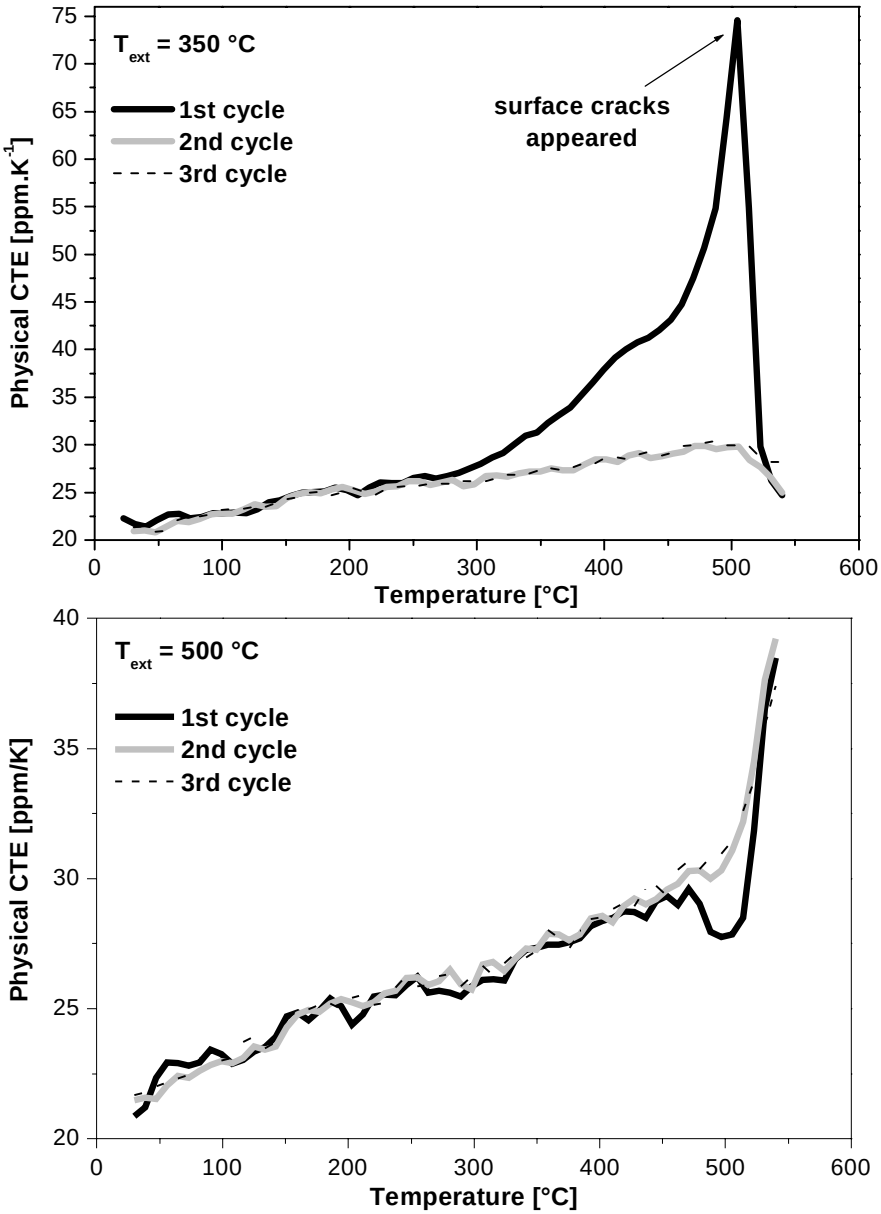

Fig. 3. CTE courses of 1 μm Al 99.7% powder compacts extruded at *R*=11:1 and different T_{ext}=350 °C (up) and 500 °C (bottom) during three subsequent heating cycles.

shape. Nevertheless massive coalescence of dispersed phase was not observed. In generally, in respect to amount of reinforcing phase within structure of compacts, detected structural stability is far superior to conventional Al-based materials.

3.2. Effect of porosity and entrapped gasses within powder compacts

Because of simple compaction without degassing and encapsulation some gases were entrapped in extruded compacts. Negative effect of entrapped gasses was in first step assessed by means of dilatometry tests. Sharp deviation from near-to-linear curve of the coefficient of thermal expansion (CTE) was observed during the first heating cycle for compacts extruded at lowest T_{ext}=350 °C (Fig. 3). This "foaming" phenomenon was clearly related to the entrapped gasses, which massively expanded during first heating cycle in closed pores and plastically deformed material. The high inner pressure led even to formation of long cracks and

Table 1. Room temperature mechanical properties (μ*HV* microhardness, yield strength R_e, ultimate tensile strength R_m, ductility A_{10}, Young's modulus E) of compacts prepared via quasi-static extrusions of 1 μm, 10 μm, and 63-400 μm Al 99.7% powders. R = 11:1.

Powder size [μm]	1				10				63-400
T_{ext} [°C]	350	400	450	500	350	400	450	500	350
μ*HV*	102.2	96.1	87.3	85.9	75.7	66.3	64.9	63.6	-
R_e [MPa]	247	238	227	213	211	183	167	160	122
R_m [MPa]	310	305	290	280	261	234	219	204	149
A_{10} [%]	9	8.7	10.9	10.4	11.1	8.4	15	4.1	13.4
E [GPa]	63.9	64.3	63	64.5	65.4	65.4	65.8	62.4	62.2

Table 2. Mechanical properties (ultimate tensile strength R_m, ductility A_{10}, Young's modulus E) of extruded Al 99.7% powder compacts obtained from tensile tests performed at 300 °C (R=11:1).

Powder size [mm]	1				10	63-400
T_{ext} [°C]	350	400	450	500	350	350
R_m [MPa]	186.7	181.7	168.9	159.9	147	79
A_{10} [%]	1.7	1.5	2.2	3.3	17.5	15.7
E [GPa]	45.7	39.2	41.3	47.2	45	48.6

to interconnection of micropores. Formed cracks enabled gas to release at testing temperature ~500 °C followed by steep CTE decline. No deviation from linear CTE was then observed at subsequent heating cycles. On the contrary, compression of gasses at higher T_{ext}=500 °C along with less work introduced into compacted powder led to rather identical "foaming" phenomena during all subsequent heating cycles with no crack formation (Fig. 3). Thermal expansion of compacts extruded at intermediate T_{ext}=400 °C and 450 °C merged somehow behaviour of two limiting cases. With increasing T_{ext} the effect of expanding gasses on CTE is observed at higher temperatures. The negative effect of "foaming" was slightly diminished by increasing of extrusion ratio to R = 20:1 and 44:1.

Compacts made of coarser 10 mm powder extruded at T_{ext}=350 °C accommodated similar "foaming" and gas release phenomenon during first heating cycle. Again break-away from linearity of CTE curve during first cycle was seen however it was shifted to higher temperature ~ 410 °C. This clearly refers to the less pronounced effect of expanding gas due to less specific powder surface and quantity of entrapped gases. Expectedly compacts prepared of the coarsest 63-400 mm powder accommodated no "gas foaming" phenomena.

3.3. Mechanical properties of powder compacts

Relatively high values of ultimate tensile strength R_m=310 MPa accompanied with quite good ductility A=9% were determined for 1 mm powder compacts extruded at 350 °C (Table 1). As the amount of work introduced during extrusion decreased (i.e. higher T_{ext}), owing to coarser microstructures R_m and μ*HV* of compacts decrease gradually. Moreover, the oxide dispersions embedded within the grain's volume additionally strengthened compacts prepared at low T_{ext}. Considerably higher strength (at roughly similar ductility A_{10}) of 1 μm powder compacts when compared to coarse powders was gained due to their finer microstructures and to higher oxide level. Further increase of plastic strain induced during extrusion (R=20:1 and 44:1) resulted in dynamic recrystallization of partial compact's regions and led to slightly deteriorated strengths (R_m = 271 MPa and 282 MPa for R = 20:1 and 44:1 respectively).

Average values of Charpy impact fracture toughness KC for all extruded compacts were at the level ~ 45 J·cm^{-2} regardless of T_{ext} and thus sufficiently fulfilled limits required by structural applications.

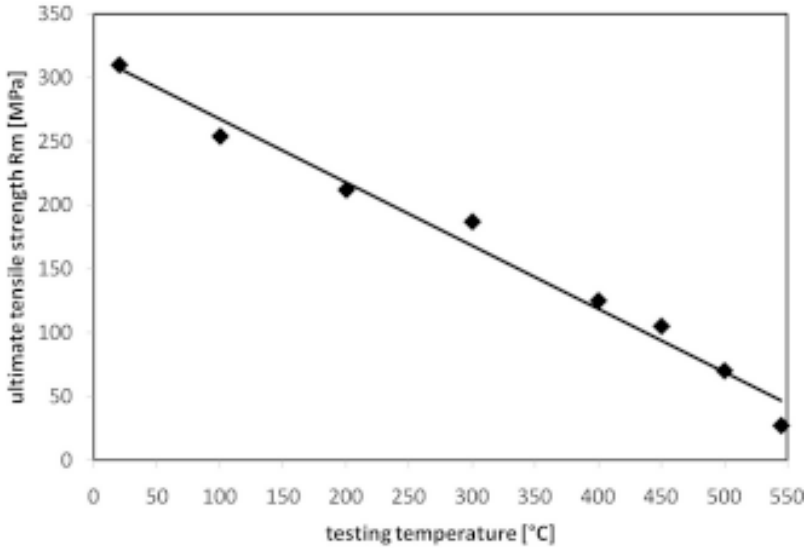

Fig. 4. Ultimate tensile strength R_m in hand with testing temperature of 1 μm Al 99.7% powder compacts (T_{ext}=350 °C, R=11:1).

Room temperature tensile tests performed on annealed 1 μm Al compacts (350 °C @ 20 hours) confirmed effective grain pinning effect of nano-scale dispersions preventing excessive grain growth at elevated temperatures. Only slight ~5% decrease in room temperature UTS after annealing was confirmed for compacts extruded at T_{ext}=350 °C. That reflected relieve of residual stresses, retained in compacts after extrusion. On the contrary 1 μm powder compacts extruded at 500 °C showed almost no alteration in room temperature R_m and A_{10} after annealing. It is seen that residual stresses concealed in compacts were proportional to amount of induced plastic work (i.e. T_{ext}). For the case of both coarser 10 mm and 63-400 μm powder compacts, thermal exposures resulted in more apparent 9% and 13% R_m declines, what was unambiguously attributed to the lack of oxide pinning spots. Annealing at extreme temperature of 500 °C @ 20 hours of 1 mm Al powder extrudates led to more significant ~16% room temperature strength decline when compared to to as-extruded state. Again higher T_{ext} with less deformation work induced responded in less absolute strength deterioration. Unlike the compacts annealed at 350 °C overall strength decline was determined predominantly by apparent grain coarsening. Ductility of all compacts after 20 hours annealing at 350 °C and 500 °C retained approximately unchanged. It should be pointed out that no blistering or cracking due to released gasses were recognized in the whole range of compacts exposed up to 500 °C.

Tensile tests performed at 300 °C revealed favourably high values of R_m ranging from 186.7 MPa to 159.9 MPa for compacts extruded in T_{ext} range 350 °C - 500 °C (R = 11:1), Table 2. With further increase of testing temperature compacts still preserved their very attractive strength and stability (Fig. 4). That reflects the importance of oxide dispersions embedded at grain boundaries regions. Besides of grain pinning effect, they are believed to act as mechanical keying barrier against grains sliding (creep behaviour improvement).

On the other hand tensile loading of compacts at 300 °C resulted in substantial, approximately three fold decrease in ductility (Table 2). Described ductility drop at elevated temperatures was governed by superimposed expanding of gasses entrapped within small pores. Small cracks, formed as a result of gasses expansion, initiated premature failure of compacts during tensile tests at 300 °C. As already the dilatometry tests had suggested (Fig. 3), expansion of entrapped gasses at 300 °C led to deterioration of high temperature ductility only if the testing temperature was close to particular T_{ext} and higher. Thus ductility of compacts at 300 °C increased from A_{10} = 1.7% to 3.3% for T_{ext} = 350

°C and 500 °C, respectively. At low T_{ext} entrapped gas was closed up and compressed in more pronounced way comparing to higher T_{ext}. Moreover pressure build-up due to decomposition of absorbed humidity from powder surfaces [7] acted in more pronounced way in case of compacts exposed to lower temperatures prior and during extrusion.

Alike the room temperature tensile tests, compacts extruded at different deformation strains (i.e. *R*) showed very similar high temperature deformation behaviour. R_m values of compacts extruded at *R* = 11:1, 20:1, and 44:1 tested at 300 °C remained approximately unchanged in the range 168.9 – 163.8 MPa. In spite of lower residual porosity reached by higher deformation rates and improved results from dilatometry tests, inferior high temperature ductility was thus improved only marginally.

Expectedly compacts of both coarser powders showed deeper relative decline in R_m determined at 300 °C (Table 2). Conversely, ductility of both coarser Al powders compacts at elevated temperatures 10 μm and 63-400 μm increased, when compared to as-extruded state. This correlates with high densities of compacts of both coarser Al powders wherein expansion of entrapped gas did not affect so distinctly their high temperature ductility.

4. CONCLUSIONS

Compacts made of ultra-fine 1 μm gas atomized Al 99.7% powder via simple and economically viable method proved its potential in application where structural stability and enhanced mechanical properties at room and mainly at elevated temperatures are of interest.

- extrusion yielded sound compacts of ultra-fine grained microstructures (transversal grain size down to 0.2 μm) strengthened with homogenously distributed nano-scopic dispersion which originated from torn surface oxide's envelopes
- promising room temperature mechanical properties of compacts were determined - high strengths up to 310 MPa accompanied with sufficient ductility ~10% and high impact Charpy fracture toughness KC ~45 Jcm^{-2}
- excellent microstructural stability was confirmed, where 20 hours exposures at 350 °C led to no structural changes and no lost of mechanical properties
- good high temperature strength (187 MPa at 300 °C), with no pronounced strength decline, was confirmed where compacts retained their relatively high strength even at testing temperatures up to 500 °C (~70 MPa)
- simple consolidation route without prior degassing resulted in the structure which contained micropores filled with gasses, entrapped and compressed during extrusion
- if the testing temperatures was about of particular T_{ext} and higher, expansion of entrapped gasses led to deterioration of high temperature ductility of compacts prepared at low extrusion ratios.

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