JOURNAL OF MATERIALS SCIENCE 26 (1991) 2208–2214

Elastic constants of $TiAl_3$ and $ZrAl_3$ single crystals

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The elastic stiffness constants, c_{ij}, were measured from the velocity of ultrasonic waves for $TiAl_3$ and $ZrAl_3$ single crystals with tetragonal $D0_{22}$ and $D0_{23}$ structures, respectively. The value of c_{11} for the $\langle 1\,0\,0 \rangle$ direction was approximately equal to that of c_{33} for the $\langle 0\,0\,1 \rangle$ direction in both $TiAl_3$ and $ZrAl_3$. Young's modulus for a single crystal was the highest in the $\langle 1\,1\,0 \rangle$ direction in which titanium or zirconium atoms and aluminium atoms were arranged in the closest packed manner, although it was not so high in $\langle 0\,1\,2 \rangle$ and $\langle 0\,1\,4 \rangle$ directions which showed the other closest packed array of the constituent atoms for $TiAl_3$ and $ZrAl_3$, respectively. The elastic constants, such as Young's modulus, shear modulus and Poisson's ratio, were approximately estimated for ideal polycrystalline $TiAl_3$ and $ZrAl_3$ from the stiffness constants and the compliance constants for single crystals. The Poisson's ratio of these materials was about 0.16 and 0.19 for $TiAl_3$ and $ZrAl_3$, respectively, and these values are much lower than those of ordinary metals and alloys. Debye temperatures were estimated at room temperature from the average velocity of ultrasonic waves and were 681 and 577 K for $TiAl_3$ and $ZrAl_3$, respectively.

1. Introduction

$TiAl_3$ and $ZrAl_3$ are intermetallic compounds with tetragonal $D0_{22}$ and $D0_{23}$ structures, respectively, and they hardly have a composition range which includes an excess or a deficiency of titanium or zirconium [1]. Each compound has a tetragonal structure which approximately consists of two or four unit cells of fcc structure, and the average interatomic distance along the c-axis is a little larger than that along the a-axis (Fig. 1 [2]). The former has a low density of $3.4\,g\,cm^{-3}$ and a melting temperature of about 1623 K, and the latter has a density of $4.1\,g\,cm^{-3}$ and a melting temperature of about 1850 K [1, 2]. They contain a large amount of aluminium and are predicted to have a remarkable oxidation resistance at high temperatures [3]. Thus, they are expected to be candidates for use in lightweight structural materials at high temperatures in the future. $MoSi_2$ and WSi_2 are also intermetallic compounds with a body centred tetragonal $C11_b$ structure and melting points of 2293 and 2437 K, respectively [1]. They are expected to be used as structural materials at temperatures up to about 1300 K. However, large single crystals of these compounds are difficult to fabricate, and the mechanical properties, such as strength and plastic deformation, have been little or never studied using single crystals for $TiAl_3$ [4] and $ZrAl_3$, respectively, although the yield strength and plastic deformation in compression at high temperatures above 1273 K were studied for $MoSi_2$ [5, 6] and WSi_2 [6].

The elastic constants of $MoSi_2$ and WSi_2, which were fundamental physical properties especially for the mechanical properties such as strength, plastic deformation and fracture, were reported previously [7] using single crystals with a diameter of 8 mm. In the present work, single crystals of $TiAl_3$ and $ZrAl_3$ were prepared using a floating zone method and the velocity of ultrasonic waves was measured at room temperature for single crystals with four orientations and their elastic stiffness and compliance coefficients were determined.

2. Experimental procedure

$TiAl_3$ and $ZrAl_3$ rods with a diameter of about 10 mm were arc-melted in an argon atmosphere using titanium and zirconium sheets which are arc-melted from sponge titanium (99%), sponge zirconium (99%), and aluminium sheets (99.99%) for preparation of single crystals. Single crystals with a diameter of 8 to 10 mm and a length of 20 to 60 mm of these intermetallics were fabricated using a floating zone method in an argon atmosphere with an optical heating furnace incorporating halogen lamps. The rate of movement of the floating zone was about 5 to $10\,mm\,h^{-1}$. Specimens for measurement of ultrasonic wave velocity

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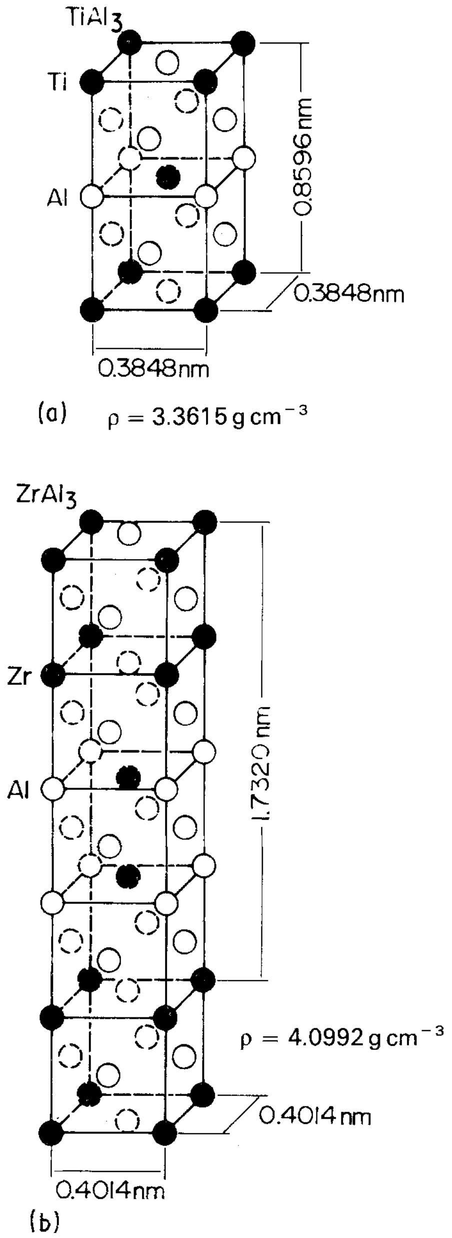

Figure 1 Crystal structures of (a) $TiAl_3$ and (b) $ZrAl_3$.

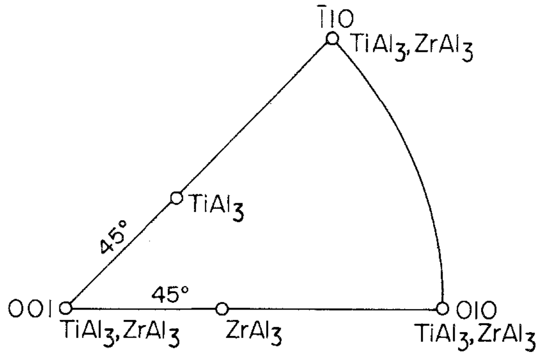

Figure 2 Crystal orientations of the plane perpendicular to the axes in which the sound wave velocity was measured.

were cut from the single crystals, and the crystal orientations of the plane perpendicular to their axes were {0 0 1}, {1 0 0}, {1 1 0}, etc., as shown in Fig. 2. The single crystals were cut using an electric discharging cutting machine with a goniometer, and the thickness of the specimens for measurements of ultrasonic wave velocity was 5 to 6 mm.

The elastic constants were calculated from the velocity of ultrasonic waves, which was measured using a simple pulse echo method. The transducers with a diameter of $\frac{1}{4}$ in. (6.35 mm) were supplied by Panametrics Co., and the resonant frequencies of 10 (model V112) and 5 (model V156) MHz were used for the longitudinal and transverse waves, respectively. The pulser/receiver was a Panametrics model 5052PR, and the oscilloscope was a YHP model 1740A (100 MHz). The couplants between the specimen and transducer were spindle oil and water-soluble couplant (Sperry Co.) for longitudinal and transverse waves, respectively. The determination of wave velocity was carried out by measuring a time interval between the first peaks of the first and the second reflection waves on the oscilloscope, because the rising point was very difficult to detect. The average values of 13 to 25 measurements were adopted for the average sound velocity.

3. Results

$TiAl_3$ and $ZrAl_3$ have tetragonal $D0_{22}$ and $D0_{23}$ structures, respectively, and the matrix of their elastic stiffness constants is given by non-zero elements of $c_{11}, c_{33}, c_{44}, c_{66}, c_{12}$ and c_{13} because of the symmetry of their crystal structure. Their elastic compliance matrix is given by the inverse matrix of their stiffness constants.

Table I shows the ultrasonic wave velocity and the associated elastic stiffness constants for various orientations of $TiAl_3$ single crystal. The sound velocities were about 8000 to 8700 m sec^{-1} for the longitudinal wave, and about 5200 to 5900 m sec^{-1} for the transverse wave. The associated elastic stiffness constants were about 2×10^2 GPa for the longitudinal wave and about 1×10^2 GPa for the transverse wave. The standard deviations were about 0.5%. The associated elastic constant for c_{44} did not change with the crystal orientation, as seen in Table I. Here, the densities of the compounds were calculated from lattice constants [2] and atomic weights of the constituent elements. Table II shows the sound velocities and the associated elastic constants for $ZrAl_3$ single crystal with various orientations. The sound velocities were about 7100 to 7700 m sec^{-1} for the longitudinal wave and about 4600 to 5000 m sec^{-1} for the transverse wave, and these values were a little lower than those of $TiAl_3$. The associated elastic constants were approximately equal to those of $TiAl_3$. The associated elastic constant from which c_{12} was obtained did not change with the thickness of the specimen. The associated elastic constant of $ZrAl_3$ corresponding to c_{44} remained constant with crystal orientation, as with $TiAl_3$. The elastic stiffness constants, c_{ij}, which were obtained from the values in Tables I and II, are shown in Table 3. The values of c_{11} and c_{33} were about 2×10^2 GPa, and those of c_{14} and c_{66} were about 1×10^2 GPa. These values were just a little higher for $TiAl_3$ than for $ZrAl_3$, and the values of c_{12} and c_{13} were the inverse. Table III also shows the elastic compliance constants, s_{ij}, which were calculated from the stiffness constants matrix, $\{c_{ij}\}$.

TABLE I The velocity of ultrasonic waves and the associated elastic constants for $TiAl_3$ single crystals

Orientation (plane)		Velocity ($m\ sec^{-1}$)		Elastic constant (10^2 GPa)	
		Average	S.D. (%)	Average	S.D. (%)
⟨0 0 1⟩	lw[a]	8046	0.37	2.175 (c_{33})	0.74
{1 0 0}	tw[b]	5237	0.23	0.922 (c_{44})	0.46
{1 1 0}	tw	5233	0.19	0.920 (c_{44})	0.38
⟨1 0 0⟩	lw	8046	0.28	2.177 (c_{11})	0.56
{0 0 1}	tw	5255	0.33	0.928 (c_{44})	0.66
{1 0 0}	tw	5888	0.23	1.165 (c_{66})	0.47
⟨1 1 0⟩	lw	8698	0.22	2.543 ($> c_{12}$[c])	0.43
Other	lw	8322	0.24	2.328 ($> c_{13}$[c])	0.49

[a] Longitudinal wave.
[b] Transverse wave.
[c] c_{12} or c_{13} is calculated from the associated elastic constant.

TABLE II The velocity of ultrasonic waves and the associated elastic constants for $ZrAl_3$ single crystals

Orientation (plane)		Velocity ($m\ sec^{-1}$)		Elastic constant (10^2 GPa)	
		Average	S.D. (%)	Average	S.D. (%)
⟨0 0 1⟩	lw[a]	7128	0.18	2.083 (c_{33})	0.36
{1 0 0}	tw[b]	4613	0.11	0.872 (c_{44})	0.23
⟨1 0 0⟩	lw	7135	0.12	2.088 (c_{11})	0.26
{0 0 1}	tw	4599	0.22	0.867 (c_{44})	0.44
{1 0 0}	tw	4994	0.14	1.022 (c_{66})	0.29
⟨1 1 0⟩ 6.16 mm thick	lw	7681	0.12	2.418 ($> c_{12}$[c])	0.23
⟨1 1 0⟩ 5.39 mm thick	lw	7680	0.12	2.418 ($> c_{12}$[c])	0.24
Other	lw	7259	0.12	2.160 ($> c_{13}$[c])	0.24

[a] Longitudinal wave.
[b] Transverse wave.
[c] c_{12} or c_{13} is calculated from the associated elastic constant.

TABLE III The elastic stiffness and compliance constants for $TiAl_3$ and $ZrAl_3$

c_{ij}	$TiAl_3$ (10^2GPa)	$ZrAl_3$ (10^2GPa)	s_{ij}	$TiAl_3$ ($10^{-3}GPa^{-1}$)	$ZrAl_3$ ($10^{-3}GPa^{-1}$)
c_{11}	2.177	2.088	s_{11}	5.075	5.568
c_{33}	2.175	2.083	s_{33}	4.939	5.235
c_{44}	0.920	0.872	s_{44}	10.870	11.468
c_{66}	1.165	1.022	s_{66}	8.584	9.785
c_{12}	0.577	0.705	s_{12}	− 1.175	− 1.663
c_{13}	0.455	0.491	s_{13}	− 0.816	− 0.920

4. Discussion

4.1. The orientation dependence of the compliance

The elastic constants such as Young's modulus and shear modulus are given for single crystals by the reciprocals of elastic compliance constants, s_{11} or s_{33}, and s_{44} or s_{66}. Thus, the orientation dependence of the Young's modulus and the shear modulus can be obtained from that of the compliance constants, s_{11} and s_{66}. The orientation dependence of s_{11} and s_{66} can be obtained from coordinate transformation of the compliance matrix $\{s_{ij}\}$. Fig. 3a and b show the orientation dependence of s_{11} for $TiAl_3$ and $ZrAl_3$ single crystals, where $1/E$ is the reciprocal of the Young's modulus which is estimated from c_{ij} and s_{ij} for isotropic materials, i.e. ideal polycrystalline materials as mentioned later. The orientation dependence of s_{11} was calculated by rotating from [1 0 0], [0 0 1] and [0 0 1] directions around [0 0 1], [1 0 0] and [$1\bar{1}0$] axes, respectively. The constant c_{11} also shows a similar behaviour to that seen in Fig. 3. The value of s_{11} for the ⟨1 1 0⟩ direction, which is the close packed direction, is smaller than that for the ⟨1 0 0⟩ or ⟨0 0 1⟩ direction. This means that the Young's modulus for the ⟨1 1 0⟩ direction is the highest in both $TiAl_3$ and $ZrAl_3$. The value of s_{11} for the ⟨0 2 1⟩ direction, which is the other close packed direction, is close to that for the ⟨0 0 1⟩ direction, and this indicates that the Young's modulus changes not only with the average interatomic distance, but also the atomic arrays in the direction of the applied stress. In general, the behaviour of s_{11} is similar for $TiAl_3$ and $ZrAl_3$, but s_{11} is much higher for the ⟨1 0 0⟩ direction than for the ⟨0 0 1⟩ and ⟨0 2 1⟩ directions for $ZrAl_3$, compared with s_{11} for $TiAl_3$. This also means that the Young's modulus changes with the atomic arrays in the direction of the applied stress.

Fig. 4a and b show the orientation dependence of s_{66} for $TiAl_3$ and $ZrAl_3$ single crystals, where $1/G$ is the reciprocal of the shear modulus which is estimated

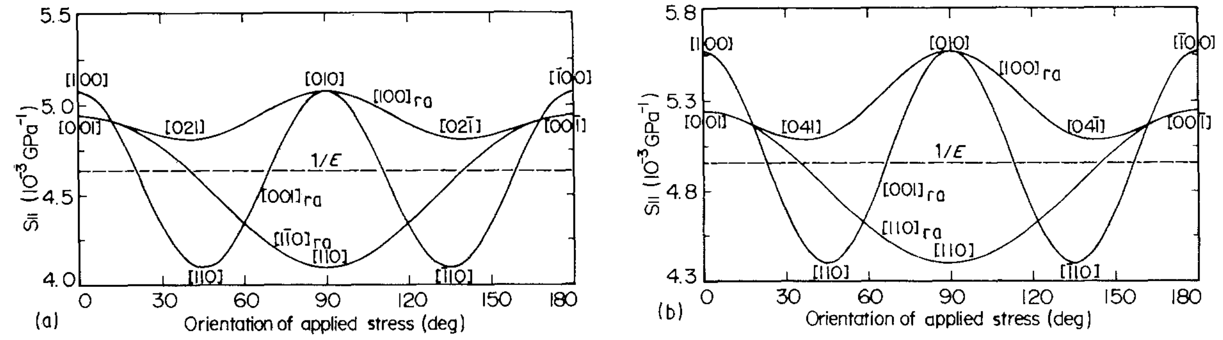

Figure 3 Orientation dependence of s_{11} for (a) $TiAl_3$, and (b) $ZrAl_3$ single crystals.

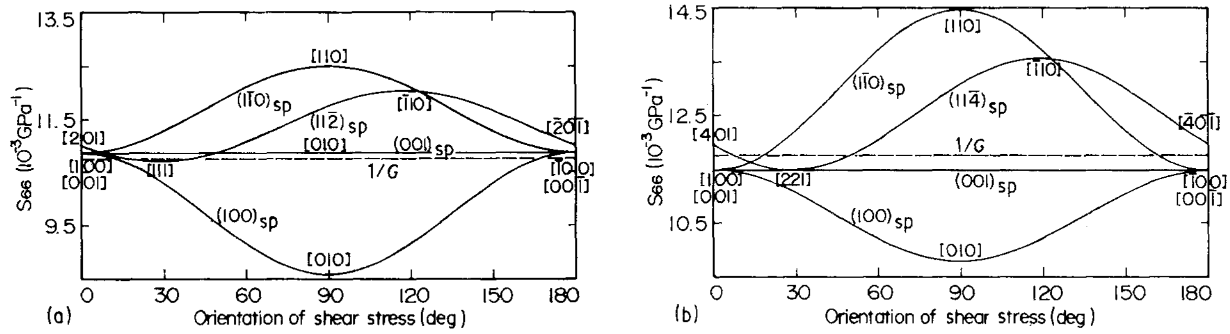

Figure 4 Orientation dependence of s_{66} for (a) $TiAl_3$, and (b) $ZrAl_3$ single crystals.

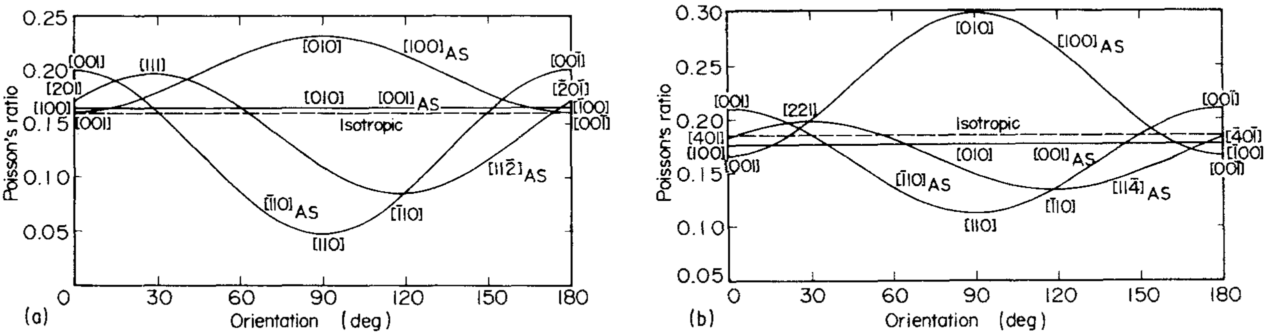

Figure 5 Orientation dependence of Poisson's ratio for (a) $TiAl_3$, and (b) $ZrAl_3$ single crystals, respectively, when the uniaxial stress is applied to some of the crystal orientations.

from c_{ij} and s_{ij} for isotropic materials, as mentioned later. The compliance s_{66} was calculated in various shear directions on various shear planes by coordinate transformation of the compliance matrix $\{s_{ij}\}$. The constant c_{66} also shows a similar behaviour to that seen in Fig. 4. s_{66} is generally large, especially in the [1 1 0] direction for both compounds, when the shear plane is (1 1̄ 0). This means that both compounds show the weakest strength elastically for shear stress in the [1 1 0] direction on the (1 1̄ 0) plane. It is relatively large in the [1̄ 1 0] direction on the (1 1 2̄) and (1 1 4̄) planes, which are close packed planes for $TiAl_3$ and $ZrAl_3$, respectively. Meanwhile, it is generally small, being smallest in the [0 1 0] direction, on the (1 0 0) plane, and this means that the shear modulus is the largest in the [0 1 0] direction on the (1 0 0) shear plane. The elastic strength is independent of the shear direction on the (0 0 1) plane. In $ZrAl_3$ the shear modulus is largely dependent on the shear direction on the (1 1̄ 0) and (1 1 4̄) planes, compared with that in $TiAl_3$.

Poisson's ratio is given by the ratio of dilatational strain, ε_{22}, perpendicular to the applied stress direction, to dilatational strain, ε_{11}, parallel to the applied stress direction when a specimen is loaded under uniaxial stress. Thus, Poisson's ratio, ν, is given by.

$$\nu = -(s_{12}/s_{22})$$

or

$$= -(s_{12}/s_{11}) \qquad (1)$$

where s_{12}, s_{11} and s_{22} are elastic compliance constants. Fig. 5a and b show the orientation dependence of Poisson's ratio for $TiAl_3$ and $ZrAl_3$ single crystals, respectively, when the uniaxial stress is applied to some of the crystal orientations. The orientation is changed around a stressed orientation. In $TiAl_3$, Poisson's ratio is the lowest in the [1 1 0] direction, which is the closest packed direction of atomic arrays, when the stress is applied to the [1̄ 1 0] direction. It is also low in the [1 1 0] direction, when the stress is applied to the [1 1 2̄] direction, although it is not low in the [1 1 1] direction, which is the other closest

TABLE IV The elastic stiffness constants of various intermetallic compounds

	c_{11} (10^2GPa)	c_{33} (10^2GPa)	c_{44} (10^2GPa)	c_{66} (10^2GPa)	c_{12} (10^2GPa)	c_{13} (10^2GPa)
$TiAl_3$	2.177	2.175	0.920	1.165	0.577	0.455
$ZrAl_3$	2.088	2.083	0.872	1.022	0.705	0.491
Ti [8]	1.624	1.807	0.467	c_{44}	0.920	0.690
Zr [8]	1.434	1.648	0.320	c_{44}	0.728	0.653
Al [8]	1.082	c_{11}	0.285	c_{44}	0.613	c_{12}
$MoSi_2$ [7]	4.170	5.145	2.042	1.936	1.042	0.838
WSi_2 [7]	4.428	5.523	2.116	2.175	1.217	0.810
Ni_3Al [9]	2.23	c_{11}	1.25	c_{44}	1.48	c_{12}
Ni_3Fe [9]	2.46	c_{11}	1.24	c_{44}	1.48	c_{12}
Cu_3Au [9]	1.87	c_{11}	0.68	c_{44}	1.35	c_{12}
NiAl [9]	2.12.	c_{11}	1.12	c_{44}	1.43	c_{12}
AgMg [9]	0.84	c_{11}	0.48	c_{44}	0.56	c_{12}
CuZn [8]	1.279	c_{11}	0.822	c_{44}	1.091	c_{12}
CuZn [9]	1.29	c_{11}	0.82	c_{44}	1.10	c_{12}
TiC [8]	5.00	c_{11}	1.75	c_{44}	1.13	c_{12}
MgO [8]	2.892	c_{11}	1.546	c_{44}	0.879	c_{12}

direction of atomic arrays. That is, elastic deformation is not easy in the [1 1 0] direction, when stress is applied in a direction perpendicular to the [1 1 0] direction. Meanwhile, it is high in the [0 1 0] direction, when the stress is applied to the [1 0 0] direction, and it remains unchanged with crystal orientation, when stress is applied to the [0 0 1] direction. Poisson's ratio for the isotropic material was calculated from the elastic constants of a single crystal, as described later. The Poisson's ratio of $ZrAl_3$ shows a similar behaviour to that of $TiAl_3$. That is, it is low in the [1 1 0] direction, which is the closest packed direction of atomic arrays, but it is not so low in the [2 2 1] direction, i.e. the other closest packed direction of atomic arrays. In both $TiAl_3$ and $ZrAl_3$, the average interatomic bonding force is expected to be strong for pulling or pushing qualitatively, when titanium or zirconium and aluminium atoms are arranged alternately in a line, compared with the case in which two types of atom are in a line of different arrangement, although the atoms are arranged in the closest packed direction. The average interatomic bonding is expected to be weak for shearing qualitatively, when two types of atom are arranged alternately in a line for the shearing direction.

The stiffness constants, c_{11} and c_{33}, are considered to be closely related to the interatomic bonding force, and then to the average interatomic distance. The ratio of the average interatomic distance in the [0 0 1] direction to that in the [1 0 0] direction is 1.117 and the ratio of c_{11} to c_{33} is 1.001 for $TiAl_3$. The former is 1.07 and the latter is 1.002 for $ZrAl_3$. These facts indicate that the elastic constants are not simply related to the average interatomic distance.

Table IV shows the elastic stiffness constants for various intermetallic compounds and elements. The stiffness constants c_{11}, c_{33}, c_{44}, and c_{66} are much higher for $TiAl_3$ and $ZrAl_3$ than for titanium, zirconium or aluminium [8] which are the constituent elements for the intermetallic compounds, and c_{12} and c_{13} are the inverse. These values for $TiAl_3$ and $ZrAl_3$ are half as large as those for $MoSi_2$ and WSi_2 reported previously [7]. The constants c_{11}, c_{33}, c_{44}, and c_{66} are approximately equal to those for Ni_3Al and NiAl [9], which are the intermetallic compounds with a cubic structure, while the constants c_{12} and c_{13} are about half the latter values, which means $TiAl_3$ and $ZrAl_3$ have lower Poisson's ratios than Ni_3Al and NiAl.

4.2. Estimation of the elastic constants for isotropic aggregates

The elastic constants, such as Young's modulus, shear modulus and Poisson's ratio, for isotropic polycrystalline materials can be estimated approximately from the stiffness constants and the compliance constants for single crystals [7, 10, 11]. The maximum values are calculated using Voigt's approximation, and the minimum values are obtained from Reuss's approximation. Hill's approximation is given by the arithmetical mean of the maximum and minimum values. Table V shows the bulk modulus, Young's modulus, shear modulus, and Poisson's ratio for ideal polycrystalline $TiAl_3$ and $ZrAl_3$, compared with those of ideal polycrystalline $MoSi_2$ [7] and WSi_2 [7]. The elastic constants of the constituent elements titanium, zirconium and aluminium [12] are also shown in the table. Both Young's modulus and the shear modulus for $TiAl_3$ and $ZrAl_3$ are half those for $MoSi_2$ and WSi_2, and Poisson's ratio for the aluminides is a little larger. Young's modulus and the shear modulus for the aluminides are more than twice those for titanium, zirconium and aluminium, and Poisson's ratio of the aluminides is about half that of the constituent metal elements. That is, the lightweight intermetallic compounds, $TiAl_3$ and $ZrAl_3$ are expected to be materials with high specific Young's modulus (the ratio of Young's modulus to the density).

The velocity of ultrasonic waves in the isotropic material is obtained from Navier's equation using the

TABLE V The elastic constants for various polycrystalline materials

	Bulk modulus (10^2GPa)	Young's modulus (10^2GPa)	Shear modulus (10^2GPa)	Poisson's ratio	Debye temperature^d (K)
$TiAl_3$^{a}	1.056	2.157	0.930	0.160	681
$ZrAl_3$^{a}	1.069	2.018	0.851	0.185	577
Ti [12]	1.072	1.08	0.401	0.345	
			0.441^b	0.319^b	380^b
Zr [12]	0.8496	0.939	0.358	0.34	
			0.364^b	0.331^b	250^b
Al [12]	0.7358	0.724	0.271	0.34	
			0.265^b	0.347^b	214^b
$MoSi_2^c$	2.097	4.397	1.911	0.151	759
WSi_2^c	2.224	4.679	2.036	0.149	625
TiC^b			1.824	0.199	1370
MgO^b			1.33	0.166	930

[a] Estimated from the elastic constants c_{ij} and s_{ij} in Table III.
[b] Estimated from the elastic constants c_{ij} and s_{ij} [8].
[c] Estimated from the elastic constants c_{ij} and s_{ij} [7].
[d] Calculated from elastic constants at room temperature.

TABLE VI The elastic wave velocity and the Debye temperature for the polycrystalline compounds

	Wave velocity (m sec^{-1})			Debye temperature, θ_D (K)
	v_l	v_t	v_m	
$TiAl_3$	8265	5260	5783	681
$ZrAl_3$	7332	4556	5022	577

density, ρ_0, the bulk modulus, K, and the shear modulus, G, shown in Table IV.

$$v_l = [(K + 4/3\,G)/\rho_0]^{1/2} \quad \text{for the longitudinal wave} \qquad (1)$$

$$v_s = (G/\rho_0)^{1/2} \quad \text{for the transverse wave} \qquad (2)$$

Then the average sound velocity, v_m, in the polycrystalline material is approximately given by [10]

$$v_m = [1/3\,(2v_s^{-3} + v_l^{-3})]^{-1/3} \qquad (3)$$

The Debye temperature, θ, is calculated from the average sound velocity, v_m, Planck constant, h, Boltzmann constant k, Avogadro constant, N, etc. [7, 10]. Table VI shows the sound velocities and Debye temperatures for ideal polycrystalline $TiAl_3$ and $ZrAl_3$. Debye temperatures for the constituent elements at room temperature are also shown in Table V. Debye temperatures for $TiAl_3$ and $ZrAl_3$ are much higher than those for titanium, zirconium and aluminium.

5. Conclusion

The elastic stiffness constants, c_{ij}, were obtained from the velocity of ultrasonic waves for $TiAl_3$ and $ZrAl_3$ single crystals with tetragonal $D0_{22}$ and $D0_{23}$ structures, respectively. The value of c_{11} for the ⟨100⟩ direction was approximately equal to that of c_{33} for the ⟨001⟩ direction in both $TiAl_3$ and $ZrAl_3$, and the ratio of c_{11} to c_{33} did not exactly correspond to the ratio of the average interatomic distance in the ⟨001⟩ direction to that in the ⟨100⟩ direction. The compliance constants, s_{ij}, were obtained from the inverse matrix of the stiffness constants matrix, $\{c_{ij}\}$. Young's modulus for a single crystal was the highest in the ⟨110⟩ direction in which titanium or zirconium atoms and aluminium atoms were arranged in the closest packed manner, although it was not so high in the ⟨012⟩ and ⟨014⟩ directions which showed the other closest packed array of the constituent atoms for $TiAl_3$ and $ZrAl_3$, respectively.

The elastic constants such as Young's modulus, shear modulus, and Poisson's ratio, were approximately estimated for ideal polycrystalline $TiAl_3$ and $ZrAl_3$ from the stiffness constants and compliance constants for single crystals. The Poisson's ratios of these materials were about 0.16 and 0.19 for $TiAl_3$ and $ZrAl_3$, and these values are much lower than those of ordinary metals and alloys. Debye temperatures were obtained at room temperature from the average velocity of ultrasonic waves and were 681 and 577 K for $TiAl_3$ and $ZrAl_3$, respectively.

Acknowledgements

The authors thank Drs K. Kawabe and T. Hirano for helpful discussions.

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Received 20 February
and accepted 9 March 1990