Dynamic and static properties of accoustic waves in gases

A QADEER, M N SHARMA AND A S VERMA

Department of Physics, University of Lucknow Lucknow 226007

The expression for the velocity of ultrasonic waves v has been modified using corrected values of the specific heat ratio γ and the empirical virial equation of state. The equation for v has been verified for a large number of polar, non-polar and mert gases at S T P and a good agreement has been obtained between the experimental and theoretical values.

The values of v thus obtained, for these gases theoretically have been utilised in computing the absorption coefficient, effective relaxation time, static and dynamic constants such as adiabatic bulk modulus and compressibility etc. of modulated high frequency sound waves.

As there is a good agreement between the calculated and experimental values of v, hence it may be presumed that the other constants computed with this value of v should also be accurate. These properties have been discussed in detail to study the molecular structure of the substances

1. INTRODUCTION

Several workers have suggested empirical and semi-empirical equations of state which describe the pressure-volume-temperature relationship of gases, liquids and liquid mixtures. It was found that none of these equations of state are in good agreement with the experimental data and hence Onnes (1901) suggested a more satisfactory and convenient form of the empirical equation. Thus equation of state can be expressed in the series form as

$$PV = RT + B'_p P + C'_p P^2 + D'_p P^3 + E'_p P^4 + \ldots \qquad \ldots \quad (1)$$

where the virial coefficients $B'_p = B_p RT$, $C'_p = C_p RT$, $D'_p = D_p RT$, and $E' = E_p RT$ are constants for a fixed temperature, but vary with temperature in a complicated manner and P is the atmospheric pressure, V is the molar volume of the gas, R is gas constant and T is the absolute temperature. These virial coefficients are also expressed in terms of powers of volume or reciprocal-pressure The virial coefficients of eq (1) can also be expressed as

$$B'_p = \frac{B'_v}{RT},$$

$$C'_p = \frac{C'_v - B'_v}{(RT)^2},$$

$$D'_p = \frac{B'_v - 3B'_v C'_v + 2B'^2_v}{(RT)^3},$$

$$E'_p = \frac{E'_v - 2C'^2_v - 4B'_v D'_v + 10B'^2_v C_v - 5B'^4_v}{(RT)^4},$$

where B'_p, C'_p, D'_v and E'_p are the virial coefficients in terms of the volume These virial coefficients may also be expressed in terms of the intermolecular functions with the help of statistical mechanics

In the present investigation, the authors have employed the simple and accurate equation of state suggested by Onnes (eq. (1)) for studying the velocity of sound v in inert, polar and non-polar gases The theoretical computed values of v have been utilised to obtain the relaxation time, the absorption coefficient and adiabatic compressibility of these gases

2. Theory

(a) *Computation of velocity of sound*

In an ideal gas the velocity of sound is generally expressed as

$$v = [\gamma^*(\partial P/\partial \rho)_T]^{\frac{1}{2}}, \qquad \qquad .. \quad (2)$$

where v is the velocity of sound in an ideal gas, γ^* the corrected value of the specific heat ratio and $(\partial P/\partial \rho)$, the variation of pressure with density of the medium Now in order to compute the values of v from eq. (2) we must know the values of the terms, γ^* and $(\partial P/\partial \rho)$ In order to compute the values of γ^* authors have developed an expression for γ^*, in terms of various thermodynamical parameters, assuming the least possible approximations The expression for γ^*, can be written as (see Appendix A)

$$\gamma^* = \gamma^0 \left[1 + p \frac{R}{C_p{}^0} \left\{ \frac{R}{C_p{}^0}(B'_p T + pC'_p T) + 2 \frac{\partial}{\partial T} \frac{(B'_p T + pC'_p T)}{1 - p^2 C_p{}'} + pC''_p \right\} \right] \qquad (3)$$

where γ^0 is the specific heat ratio of the gas at 0°C. C^0_v, C^0_p are respectively the specific heats at constant volume and constant pressure at 0°C, B'_p, C'_p are constants related to second and third virial coefficients. p is the pressure in atmospheres and T is the absolute temperature

Now eq. (1) can be expressed in terms of density in as

$$PM/\rho = RT + B'_p P + C'_p P^2, \qquad \qquad ... \quad (4)$$

where $M/\rho = V$.

Differentiating eq. (4) with respect to ρ and keeping T constant we get the value of $(\partial P/\partial \rho)$. Substituting the value of $(\partial P/\partial \rho)$. Eq (2) yields

$$v = \left[\gamma^* \frac{(RT + 2B'_p P + 3C'_p P^2)}{M} \right]^{1/2}. \tag{5}$$

The theoretical value of v computed from eq (5) have been presented in table 1, where they have been compared with the experimental values.

Table 1   Values of the velocity of sound at 0° C

		Velocity of sound v in meter/sec		
		Observed	Theoretical eq (5)	
(A)	Inert gases			
	Helium	970 0[a]	969 7	0 09
	Neon	435 0[a]	429.5	0 11
	Argon	319 0[a]	317.9	0 31
	Krypton	—	212 7	
	Xenon	—	169 7	
(B)	Non-polar gases			
	Hydrogen	1269 5[a]	1262.3	0 58
	Nitrogen	337 0[a]	337.4	0 13
	Oxygen	317.2[a]	315 3	0 60
(C)	Polar gases			
	Ammonia	415 0[b]	415 9	0 21
	Carbondioxide	258.0[b]	260 0	0 79
	Carbonmonoxide	337 1[b]	338 0	0.28
	Chlorine	205.3[b]	204 4	0 42
	Nitric oxide	325 0[b]	326 0	0 29
	Nitrous oxide	261.8[b]	260 7	0.44
	Sulpherdioxide	213 0[b]	212 1	0 40
	Ethane	308 0[b] (10°C)	304 9	0 90
	Ethylene	317.0[b]	318 6	0.50
	Methane	430 0[b]	430 7	0 16

[a]Grey (1957)
[b]Hand Book of Physics and Chemistry (1962).

(b) *Relaxation phenomena in gases*

When the ultrasonic waves pass through a fluid it takes a finite time for the medium to attain the equilibrium due to the relaxation of the molecules of the fluid. The thermal energy of molecules of a system is, in general made up of translational, rotational and vibrational energy which are normally in equilibrium. The adiabatic compression of the gas due to the passage of a high

frequency sound wave, first increases the translational energy only, but as a result of collisions, part of that energy becomes converted into internal energy rotational and possibly vibrational. Equilibrium between the three states is restored gradually, giving rise to the relaxation time of the system   Considerable work has been done on the computation of relaxation time in the case of fluids.   Bhatia (1967) has given a detailed study of this phenomena from the quantum mechanical point of view which is too cumbersome and has a very little improvement over the classical theory as pointed out by Bhatia (1967) and Nozdrev (1960).   However, in the present work, the authors have presented a very simple treatment for the evaluation of relaxation time from classical point of view

The relaxation time τ mainly depends on viscosity (η) and thermal conductivity (K) (Bhatia 1967).   However, τ also depends upon rotational and vibrational collision of molecules but their contribution is negligibly small (about ± 2 to 3%) Stokes (1845) assumed that the dependence of τ on η can be expressed as

$$\tau_\eta = \frac{4}{3} \cdot \frac{\eta}{cv^2} , \qquad \qquad \cdots \ (6)$$

where ρ is the density of the medium   Later on Kirchoff (1968) proposed that τ also depends on thermal conductivity (K) of the medium and can be written as

$$\tau_{th} = \frac{K(\gamma^* - 1)}{\rho C_p v^2} , \qquad \qquad \cdots \ (7)$$

where C is the specific heat of the fluid at constant pressure

The effective values of τ due to η and K can be expressed as

$$\frac{1}{\tau_{\rho 66}} = \frac{1}{\tau_\eta} + \frac{1}{\tau_{th}}$$

or $\qquad \qquad \qquad \qquad \qquad \qquad \qquad \qquad \qquad \qquad \cdots \ (8)$

$$\tau_{\rho 66} = \frac{\tau_\eta \tau_{th}}{\tau_\eta + \tau_{th}}$$

The computed values of τ_η from eq. (6) τ_{th} employing eq (7) and τ_{66} using eq. (8) are presented in table 2.

(c) *Classical absorption coefficient of sound*

Stokes (1845) and Kirchoff (1968) have successfully shown that when a high frequency sound wave passes through a gaseous medium, the absorption of quanta of phonons takes place, the rate of absorption per cm² is known as the absorption coefficient.   Like relaxation time the classical absorption coefficient

Table 2. Values of classical absorption coefficient, effective relaxation time, static and dynamic constants for gases at STP*

Gases	$\frac{\alpha_\eta}{f^2} \times 10^{13}$ nep.cm⁻¹ sec⁻²	$\frac{\alpha_{th}}{f^2} \times 10^{13}$ nep.cm⁻¹ sec⁻²	$\frac{\alpha_{class}}{f^2} \times 10^{13}$ nep.cm⁻¹ sec⁻²	$\tau_\eta \times 10^{10}$ sec.	$\tau_{th} \times 10^{10}$ sec.	$\tau_{obs} \times 10^{10}$ sec.	$\frac{\alpha_{th}/f^2}{\alpha_\eta/f^2}$	$\frac{\tau_{th}}{\tau_\eta}$	$K' \times 10^{-6}$ Dyne cm⁻¹sec⁻²	$B \times 10^7$ Dyne⁻¹ cm.sec⁻²	η_0 centipoise
(A) Inert											
Helium	0·316	0·220	0·536	1·548	1·082	0·637	0·612	0·614	1 678	5 690	0·011
Neon	1·150	—	—	2·500	—	—	—	—	1 660	6 020	—
Argon	1·125	0·783	1·908	1·750	1·220	0·726	0·684	0·691	1 694	5·093	0·012
Krypton	1·802	—	—	1·948	—	—	—	—	1 693	5·906	—
Xenon	2·054	—	—	1·674	—	—	—	—	1 698	5 890	—
(B) Non-Polar											
Hydrogen	0·129	0·055	0·184	0·824	0·353	0·247	0·424	0·424	1 427	7 008	0·034
Nitrogen	0·951	0·380	1·331	1·639	0·648	0·460	0·399	0·400	1·424	7 002	0·062
Oxygen	1·193	0·465	1·658	1·905	0·743	0·535	0·389	0·381	1 421	7 03	0·076
(C) Polar											
Ammonia	0·464	0·101	0·565	0·976	0·213	0·174	0·218	0·218	1 331	7·501	0·002
Carbon dioxide	1 118	0·286	1 404	1 467	0·376	0·299	0·256	0·256	1 329	7·525	0·003
Carbon monoxide	0·961	0·345	1 306	1 641	0·588	0·433	0·359	0·355	1 422	7·030	0·006
Nitrous oxide	1 082	0·365	1 447	1 288	0·385	0·297	0·300	0·300	1 377	7·261	0·004
Nitric oxide	1 113	0·298	1 411	1 783	0·601	0·467	0·337	0·337	1 420	7 044	0·007
Chlorine	1 229	0·368	1 597	1 460	0·391	0·309	0·268	0·268	1 326	7 540	0·004
Sulpher-dioxide	0·805	0·243	1 048	0·871	0·263	0·202	0·301	0·301	1 339	7 469	0·003
Ethane	0·632	0·128	1·760	0·971	0·197	0·164	0·202	0·202	1·249	8 005	0·002
Ethylene	0·641	0·134	0·775	1 038	0·217	0·179	0·209	0·208	1·286	7·775	0·002
Methane	0·502	0·145	0·647	1 093	0·316	0·245	0·289	0·289	1 329	7·526	0·003

* The required data in present investigation have been taken from Grey (1972).

(α_{class}) also mainly depends on η and K of the gases and may be simply written as (Bhatia 1967)

$$\frac{\alpha_{class}}{f^2} = \frac{\alpha_n}{f^2} + \frac{\alpha_{th}}{f^2} \qquad \ldots (9)$$

in which

$$\frac{\alpha_n}{f^2} = \frac{2}{3} \frac{\omega^2 \eta}{f^2 \rho v^3} \qquad \ldots (10)$$

and

$$\frac{\alpha_{th}}{f^2} = \frac{\omega^2 K (\gamma^* - 1)}{2\rho f^2 v^3 C_p} \qquad \ldots (11)$$

where ω is the cyclic frequency of the generator and $W = 2\pi f$, f being the frequency

Now substituting the values of (α_n/f^2) and (α_{th}/f^2) from eqs. (10) and (11) respectively in eq (9) we at once get

$$\frac{\alpha_{class}}{f^2} = \frac{2}{3} \frac{\omega^2 \eta}{f^3 \rho v^3} + \frac{\omega^2 K (\gamma^* - 1)}{2\rho f^2 v^3 C_p} \qquad \ldots (12)$$

The calculated values of α_n/f^2, α_{th}/f^2 and α_{class}/f^2 employing eq (10), (11) and (12) respectively for eighteen gases are tabulated kn table 2

(d) *Relation between* τ_{th}/τ_η *and* $\dfrac{\alpha_{th}/f^2}{\alpha_n/f^2}$

It is important to mention here that τ_{p66} and α_{class}/f^2 mainly depend on η and K of the fluids. Therefore this suggests that there should be some direct relationship between the terms involving τ_{p66} and α_{class}/f^2 For the present need, we devide eq (7) by eq (8) and eq (11) by eq (12) and we find a simple relation between the relaxation times and absorption coefficients as

$$\frac{\alpha_{th}/f^2}{\alpha_n/f^2} = \frac{\tau_{th}}{\tau_\eta} \qquad \ldots (13)$$

From eq (13) it is clear that if we know any three quantities the fourth one can be easily evaluated Moreover eq (13) can be interpreted as *the ratio of the absorption coefficient of sound waves due to thermal conduction and that due to viscosity is the same as the ratio of the relaxation time due to thermal conduction and viscosity.*

The values of the ratio τ_{th}/τ_η and $\dfrac{\tau_{th}/f^2}{\alpha_\eta/f^2}$ have been given in table 2 (columns III and IV respectively.

(e) *Elastic constants*

From the knowledge of velocity of sound we can also evaluate the values of elastic constants. In the present investigation, we have theoretically computed the adiabatic bulk modulus of elasticity (K') and compressibility (β) employing the following expressions

$$K' = v^2 \rho \qquad \qquad \dots (14)$$

and

$$\beta = \frac{1}{v^2 \rho} \qquad \qquad \dots (15)$$

The theoretical values of K' and ρ are presented in table 2. The bulk viscosity η is the ratio of the effective relaxation time $(\tau_{\rho 66})$ and compressibility (β). Mathematically we can express as (Bhatia 1967)

$$\eta = \frac{\rho v v}{\beta} \qquad \qquad \dots (16)$$

Substituting the values of $\tau_{\rho 66}$ from eq (8) and from eq. (15), eq (16) yields

$$\eta_0 = \frac{4K\eta(\gamma^* - 1)}{4\eta C_p + 3K(\gamma^* - 1)} . \qquad \qquad \dots (17)$$

The computed values of η_0 from eq (17) are also reported in table 2

3. Results and Discussion

In calculating the velocity of sound in gases using eq. (5) two important points have been taken into account viz., (a) the equation of state has been utilised upto the third virial coefficient and (b) in eq (2) values of γ^* calculated at $0°C$ have been employed. It is obvious from table 1 that the theoretical values of v are in close agreement wigh the experimental values within the limits of experimental error. Thus the theoretical values of v are satisfactorily reliable and these can be employed for evaluating allied properties of these gases such as relaxation time, absorption coefficient etc

The values of τ_η and τ_{th} calculated on the basis of eqs. (6) and (7) respectively are presented in table 2. Assuming that the classical relation time mainly depends on τ_η and τ_{th}, and neglecting the intramolecular collisions the values of τ_{class} have been computed and are also given in the same table. The effect of viscosity of relaxation time is found to be greater than the effect of thermal conductivity i.e., $\eta_\eta > \eta_{th}$. This may be due to the fact that for gases the values of K is almost neglegible and hence the effect of η predominates on the τ values. On the basis of the theoretical results obtained in the present work we can safely

conclude that τ depends on the nature of bonding between two electrons of the atom. More the number of bonds, shorter is the relaxation time viz , as molecules are tightly packed, the binding force is greater and therefore the values of relaxation time becomes small   For example, carbon monoxide and nitric oxide having triple bond have larger relaxation time. Carbon dioxide, nitrious oxide, sulpher dioxide and ethylene with double bond have smaller relaxation time than carbon monoxide and nitric oxide having triple bonds   The other reason which we can assign to this is that carbon monoxide and nitric oxide is more polar than carbon dioxide, nitrous oxide etc.   Moreover, τ for carbon monoxide $> \tau$ for carbon dioxide, because carbon monoxide has two unpaired electrons while CO_2 has only one unpaired electron.   Hence we can also say that values of τ also depends on the number of unpaired electrons viz , τ increases as the number of unpaired electrons increases.   The saturated vapours such as ammonia, ethane and methane have the smallest value of relaxation time.   On the other hand the bondless inert gases have the largest value of relaxation time $(0 > 1 > 2 > 3 > 4)$ according to the bond number).

The values of α_n/f^2, α_{th}/f^2 and α_{class}/f^2 are tabulated in table 2   Generally the total absorption coefficient is expressed as

$$\frac{\alpha_{total}}{f^2} = \frac{\alpha_{class}}{f^2} + \frac{\alpha_{kin}}{f^2}$$

because of non-availability of the data for the term (α_{kin}/f^2) and also since the contribution of (α_{kin}/f^2) is usually very small being of the order of 1 to 5%, we have neglected the contribution of this term in the computation of (α_{total}/f^2)

$$\frac{\alpha_{total}}{f^2} \simeq \frac{\alpha_{class}}{f^2}$$

It is seen from the table that for all the gases studied $(\alpha_{th}/f^2) < (\alpha_n/f^2)$ The maximum value of α_{th}/f^2 is 36% in the case of CO and NO and for NH_3 the maximum value is 78 1%

The $(\alpha_{th}/f^2) < (\alpha_n/f^2)$ for all the gases investigated in the present work   The maximum value of (α_{th}/f^2) is about 40% in the case of helium and the minimum value is about 27 2% for nitrogen (α_n/f^2) has the maximum value for nitrogen about 72.8% and it is minimum for helium (60%).   The nature of variation of (α_{th}/f^2) and (α_n/f^2) is the same as that of τ_{th} and τ_n which is in accordance with the molecular theory of gases

The theoretical values of the elastic constants K' and β obtained from eqs (14) and (15) respectively in conjunction with eq (5) for v are presented in table 2 The bulk viscosity η_0 computed on the basis of eq. (17) are also tabulated in table 2.   It is interesting to mention here that like τ, η_0 also depends on the

number of bonds. Hence we can frame a rule to arrange these gases on the basis of the values of η_0 as

$$A, CO, NO > CO, CO_2, NO_2, SO_2, C_2H_4 \sim NH_3, C_2H_6, CH_4$$
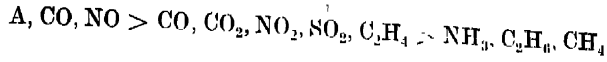
APPENDIX A

The virial equation of state for second order of pressure (P) is written as

$$PV = RT + B'_p P + C'_p P^2 \qquad \text{(A1)}$$

From eq. (A1) we can write

$$V = \frac{RT}{P} + B'_p P + C'_p P^2 \qquad \text{(A2)}$$

and

$$P = -\frac{RT}{V} + B'_p P + C'_p P^2 \qquad \text{(A3)}$$

Differentiating eq. (A2) with respect to p keeping T constant and differentiating eq. (A3) with respect to V keeping T constant, we get

$$\left(\frac{\partial V}{\partial p}\right)_T = -RT \frac{(1 - p^2 C')}{p^2} \qquad \text{(A4)}$$

and

$$\left(\frac{\partial p}{\partial V}\right)_T = -\frac{p^2}{RT} \qquad \text{(A5)}$$

Now we know that

$$-\frac{1}{V^2}\left(\frac{\partial V}{\partial p}\right)_T = \frac{1}{M}\left(\frac{\partial P}{\partial \rho}\right)_T . \qquad \text{(A6)}$$

Substituting the values of V from eq (A2) and $(\partial V/\partial p)_T$ from eq (A4) in eq. (A6) we get

$$\left(\frac{\partial p}{\partial \rho}\right)_T = \frac{RT}{M}\left(\frac{1 + B'_p P + C'_p P^2}{1 - p^2 C_p}\right) \qquad \text{(A7)}$$

From thermodynamics we know that

$$\left(\frac{\partial H}{\partial T}\right)_p = C_p, \qquad \text{(A8)}$$

and

$$\left(\frac{\partial H}{\partial p}\right)_T = -T\left(\frac{\partial V}{\partial T}\right)_p + V \qquad \text{(A9)}$$

Transformation of relations (A8) and (A9) yields

$$\frac{\partial}{\partial p} C_p = -T\left(\frac{\partial^2 V}{\partial T^2}\right)_p \qquad \text{(A10)}$$

Substitution of the value of V from eq (A2), (A10) yields

$$\frac{\partial}{\partial p} C_p = -RT \left[\frac{\partial^2}{\partial^2 T^2} \{(B'_p T) + P(C'_p T)\} \right] \qquad \dots \text{(A11)}$$

Integrating eq. (A11), we get

$$C_p = C^0_p - pRT \frac{\partial^2}{\partial T^2} \{(B'_p T) + p(C'_p T)\}. \qquad \dots \text{(A12)}$$

Now differentiating eq (A2) with respect to T, keeping P constant

$$\left(\frac{\partial V}{\partial T} \right)_p = \frac{\partial}{\partial T} \left[RT + \left(\frac{1}{p} + B'_p + C'_p P \right)^2 \right]$$

$$= \frac{R}{p} \left[1 + p \frac{\partial}{\partial T} \left\{ B'_p T + p^2 \frac{\partial}{\partial T} (C'_p T) \right\} \right] \qquad \dots \text{(A13)}$$

$$\therefore \quad (C_p - C_V) = -TV \left(\frac{\partial V}{\partial T} \right)_p \frac{1}{V} \left(\frac{\partial p}{\partial V} \right)_T$$

$$\therefore \quad (C_p - C_V) = -T \left(\frac{\partial V}{\partial T} \right)_p^2 \left(\frac{\partial p}{\partial V} \right). \qquad \dots \text{(A14)}$$

Substituting eqs. (A5) and (A13) in eq (A14), we get

$$(C_p - C_V) = R \left[1 + 2p \frac{\partial}{\partial T} (B'_p T) + 2p \frac{\partial^2}{\partial T^2} (C'_p T) \right] \qquad \dots \text{(A15)}$$

We can write

$$C_v = C_p - (C_p - C_v) \qquad \dots \text{(A16)}$$

Putting the values of C_p and $(C_p - C_V)$ from eqs. (A12) and (A15) respectively in eq. (A16), one gets

$$C_v = C^0_v - pR \left[T \frac{\partial^2}{\partial T^2} \left\{ (B'_p T) + p(C'_p T) + \frac{\partial}{\partial T}(B' T) + \frac{2p \frac{\partial}{\partial T}(C'_p T)}{1 - p^2 C'_p)} \right\} \right]. \qquad \dots \text{(A17)}$$

Dividing eq. (A11) by eq. (A17), we get

$$\frac{C_p}{C_v} = \frac{C_p{}^0 - pRT \frac{\partial^2}{\partial T^2} \{B_p' T + p(C_p' T)^2\}}{C_v{}^0 - pR \left[T \frac{\partial^2}{\partial T^2} \{B_p' T + p(C_p' T)\} + \frac{2\partial}{\partial T} \frac{B_p' T + 2p \, \partial/\partial T(C_p' T)}{1 - p^2 C_p'} \right]} \qquad \dots \text{(A18)}$$

Substituting $\dfrac{C_p{}^0}{C_v{}^0} = \gamma^0$ and $\dfrac{C_p}{C_v} = \gamma^*$ in eq. (A18) and solution of it yield

$$\gamma^* = \gamma^0[1 + pRT \frac{\partial^2}{\partial T^2}(B_p{}'T) + p(C_p{}'T)\left(\frac{C_p{}^0 - C_v{}^0}{C_p{}^0 C_v{}^0}\right) + 2p\frac{R}{C_v{}^0}\frac{\partial}{\partial T}(B_p{}'T + pC_p{}'T).$$

$$\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad \dots \quad (A19)$$

$$C_p{}^0 - C_v{}^0 = R^0 = R \text{ (say)},$$

hence,

$$\gamma^* = \gamma^0\left[1 + \frac{p}{C_v{}^0}\left\{\frac{R}{C_p{}^0}\left[T\frac{\partial^2}{\partial T^2}(B'_p T + pC_p{}'T) + 2\frac{\partial}{\partial T}\frac{B_p{}'T + pC_v{}'T}{1 - p^2 C_p{}'}\right]\right\}\right].$$

$$\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad\qquad \dots \quad (A20)$$

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